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# *Strategy and Methodology for Radioactive Waste Characterization*



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## FOREWORD

Radioactive waste management requires planned and systematic actions to provide confidence that the entire system, processes and final products will satisfy given requirements for quality. In order to ensure a quality end product, it is absolutely necessary to know and control the chemical and radiochemical parameters within the entire waste management life cycle, with special emphasis on waste conditioning, storage and disposal. Testing and analyses to demonstrate the radioactive content and the quality of final waste forms and waste packages are key components of this knowledge and control and are essential to accurate characterization of the waste.

Reliable and effective methods for measuring radiological, physical, chemical, radiochemical and other characteristics have been established in many Member States, and the IAEA has several technical reports series on the subject. However, none of the existing IAEA publications has attempted to present a strategy and methodology for waste characterization which takes into account the origin of the waste, the different waste streams, the situation of the analytical laboratories, and the analytical technologies and techniques available. Neither has any attempt been made to offer any selection of standards that are used in the radio-analysis laboratories. To cover this gap, the IAEA initiated a task that was intended to give recommendations on harmonization in waste characterization. During drafting, the scope was appropriately expanded to provide a more comprehensive examination of strategies for radioactive waste characterization.

The preparation of the report was accomplished through two consultant meetings in November 2002 and March 2004, along with one technical meeting held in March 2003. A final comprehensive review was performed August–September 2006 to verify the information remained current and relevant prior to publication. In total, 19 experts from 13 Member States and the IAEA participated at different stages in the development process.

The IAEA wishes to express its appreciation to all those individuals who took part in the preparation and publication of this report. Particular acknowledgement is due to P. Van Iseghem, Belgium, who chaired both consultants meetings and the technical meeting, as well as contributing great effort toward the completion and technical review.

The officer at the IAEA responsible for initiating this report was R. Burcl of the Division of Nuclear Fuel Cycle and Waste Technology. J.L. González Gómez and J.J. Kelly of the same division finalized the report for publication.

### *EDITORIAL NOTE*

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## SUMMARY

Over the past several decades, there has been an increasing emphasis on the importance of producing a 'quality end product' for all disposed radioactive waste. This is achievable only by obtaining a thorough and accurate assessment of the physical, chemical and radiological characteristics of the waste, a process which is referred to as 'waste characterization.' This must be accomplished or verified at the point of generation, during waste conditioning, and upon disposal, with a clear demonstration that the waste meets the performance objectives established by the disposal acceptance criteria. Moreover, it must be accomplished in a systematic manner using proven methodologies, technologies and techniques with an overriding emphasis on quality assurance and quality control.

The existing literature identifies and discusses reliable and effective methods for measuring radiological, physical, chemical, radiochemical and other characteristics of radioactive waste. An examination of the waste characterization strategies and practices in various countries indicates that many different or modified control methods are in use. Insufficient harmonization of testing methods and different interpretation of the obtained results, especially in the area of quality control of the final waste forms and waste packages, constrain application of unified safety rules and requirements.

The IAEA has also produced several Technical Reports Series publications on the subject of waste characterization. This publication goes beyond the existing literature by providing a comprehensive examination of the methodologies and strategies for waste characterization which takes into account the origin of the waste, the different waste streams, the situation of the characterization laboratories, and the analytical technologies and techniques available. It provides waste generators, processors, laboratories, and other involved organizations with guidelines on establishing a radioactive waste characterization quality control strategy and policy. It also provides comprehensive information on the available quality control procedures used in the various steps of waste processing, their critical evaluation, and suggested optimum control systems/methods for enhancing the quality and accuracy of waste characterization. A review of the recent state of standardization activities is also included, as well as information for harmonization of testing procedures in various countries.

This publication provides:

- A review of the requirements for and development of a waste characterization programme strategy, quality assurance programme, and quality control activities at the waste generator, processor, repository, and local or national laboratory levels.
- A review of characterization responsibilities applicable to waste generators, processors, and repository operators, as well as an examination of the cost and benefits of waste characterization.
- A review of the important factors to be considered in a waste characterization programme, including accuracy and uncertainties, scaling factors, and measurement methods.

- A discussion of the applicability of various waste characterization methodologies to specific categories of waste streams (simple/stable waste streams, complex/variable waste streams, decommissioning waste streams, etc.).
- A discussion and a tabulated review of the most commonly used characterization methods and techniques.

Over the past decade significant progress has been achieved in the development of waste characterization and control procedures and equipment as a direct response to ever-increasing requirements for quality and reliability of information on waste characteristics. Failure in control procedures at any step can have important, adverse consequences. In some cases, errors in waste characterization may result in producing waste packages which are not compliant with the waste acceptance criteria for disposal, thereby adversely impacting the repository. The information and guidance included in this publication corresponds to recent achievements and reflects the optimum approaches, thereby reducing the potential for error and enhancing the quality of the end product.

# 1. INTRODUCTION

## 1.1. BACKGROUND

Proper control of chemical and radiochemical parameters of radioactive waste within the entire waste management life cycle, and careful testing of the quality of final waste forms and waste packages, are principal components in any waste management strategy. Failure in control procedures at any step can cause important consequences, not only in follow-up steps, but, in some cases, may result in generating waste packages which are not compliant with the waste acceptance criteria for long term storage or disposal.

In the past decade significant progress has been achieved in the development of waste characterization and control procedures and equipment as a direct response to ever-increasing requirements for quality and reliability of information on waste characteristics. A good reference describing the components of the waste characterization process has been published by the Agency [1].

Analyzing the situation in various countries (and even in particular organizations or laboratories dealing with radioactive waste within the country), it can be recognised that many different or modified, control methods are used. Insufficient harmonization of testing methods and different interpretation of obtained results, especially in the area of quality control of final waste forms and waste packages, constrain application of the unified safety rules and requirements. Although reference [1] describes the components of waste characterization, it does not describe the strategic considerations and guidance for establishing and maintaining a good waste characterization programme. This publication provides guidance to establish a reliable waste control strategy and to advise on the selection of verified chemical and radiochemical methods and procedures applicable in any particular step in the waste management life cycle.

## 1.2. OBJECTIVE

The objectives are:

- Establish a process for characterising radioactive waste during its entire life cycle;
- Contribute to safety during the different stages of the radioactive waste life cycle: generation, processing, transport, storage and disposal;
- Demonstrate that the acceptance criteria (reception, processing, transport, storage, disposal) are met based on the performance assessment for the disposal concept; and
- Provide technical input to:
  - Identify the necessary and sufficient (adequate and cost effective) characterization requirements;
  - Identify the accuracy requirements (of particular interest to the regulator);
  - Categorise the waste, and identify the associated regulatory requirements (needs, selection processing);
  - Identify the quality assurance needs and related actions;
  - Contribute to the selection of the optimum waste forms (in view of disposal); and
  - Demonstrate that the candidate waste forms meet the specifications.

The primary purpose of this publication is to provide waste control laboratories, waste generators, waste conditioners, and other involved organizations with guidelines on establishing radioactive waste characterization quality control strategy, policy and methodology. Another purpose is to offer comprehensive information on available quality control procedures used in the various steps of waste processing, their critical evaluation, and suggested optimum control systems/methods. These correspond to recent achievements and reflect in the best way the requirements of each particular user. A review of the recent state of standardization activities is also included, as well as information for harmonization of testing procedures in various countries.

### 1.3. SCOPE

During the drafting process, available information and experiences from various waste generators, waste processing organizations, and waste control laboratories were collected and reviewed by an international team of experts who evaluated advantages and disadvantages and proposed potential ways to increase the harmonization of control procedures. The strategy of radioactive waste control during its entire life-cycle and the requirements for characterization of waste forms and waste packages for long term storage and disposal are other subjects covered in the report. The status of normative documentation and standardization activities are also discussed.

The publication consists of two principal themes: the first dealing with waste characterization and control strategy, and the second providing a description and evaluation of selected control procedures and methods for all basic steps of the radioactive waste life cycle. The following issues are covered:

- Requirements (general) for raw waste characterization, quality control of waste processing technologies, and quality control of conditioned waste (for storage and disposal);
- Strategy of waste control (for all steps mentioned above);
- Selected control procedures, their evaluation, and proposal of the most convenient methods for characterization of various kinds of raw waste;
- Selected control procedures, their evaluation, and proposal of the most convenient methods for monitoring and control of waste processing technologies;
- Selected control procedures, their evaluation, and proposal of the most convenient methods for destructive and non-destructive control of waste forms and waste packages;
- List of relevant norms and standards;
- Status of unification of control methods and standardization activities; and
- Potential options and recommendations for further harmonization of control procedures.

### 1.4. STRUCTURE

The publication is composed of four technical sections and a large set of annexes. Section 2 describes how a characterization programme should be defined. It discusses the elements, organisation and responsibilities evaluating the costs and benefits of such a programme. Scaling factors and accuracy of measurements and its consequences are the core of Section 3. With regard to the accuracy of the measurements, a large part of Section 3 is devoted to a possible interlaboratory comparison and to the calculations of the uncertainties.

In Section 4, several methodologies are discussed, depending on the characteristics and origin of different wastes streams. It includes simple, complex, stable and variable streams, and the methodologies for characterization of decommissioning and “historic” wastes.

Section 5 introduces destructive and non-destructive assays and some of the equipment to be used. For different methods, gamma spectroscopy, neutron assay, or analytical methods, an overview is provided of the most important characteristics and usefulness of specific assays. Finally, the annexes include descriptive parts, such as gamma scanning, relevant reports and standards, and an example calculation of uncertainties when dealing with samples assumed to be representative.

## **2. STRATEGY IN DEFINING THE CHARACTERIZATION PROGRAMME**

### **2.1. BACKGROUND**

Characterization actions are currently being performed during various stages of the radioactive waste life cycle: during generation (including waste retrieval activities), processing (treatment, conditioning), and storage/disposal. The type and extent of the characterization efforts depend on many factors, e.g.:

- the type of waste or waste form;
- the disposal concept;
- the regulatory regime;
- the amount of process knowledge that is available;
- the kind of characteristics to be measured.

It is consequently impossible to define or recommend a single characterization procedure for even similar waste forms or packages. In order to elaborate a strategy for the characterization actions in any given case, the following should be critically understood and assessed:

- the waste acceptance criteria (and the rationale behind it); and
- the safety assessment for the disposal concept (e.g. assumptions made, information needed).

It is of importance that waste characterization is considered as part of the larger waste management strategy, and interactions should be occurring among the various working groups involved. One of the outputs of this is that the minimum accuracy required and detection limits of the characterization efforts may be better identified.

Defining the organisational scheme into which the characterization efforts fit will take into account the relationships between the various bodies involved: those responsible for waste management, the disposal organisation, the regulator, the various waste producers and operators, and the independent laboratory. A basic quality principle relates to the independence of certain characterization facilities and activities:

- The generator of the radioactive waste and the operators (treatment and conditioning, storage and disposal) perform characterization programmes for various reasons.
- These activities need to relate with and report to the waste management staff. The independent laboratory must respond and report to the regulator.
- The application of an additional quality control (independent laboratory) is a basis of quality assurance and will strongly contribute to the public confidence in the back-end of the nuclear fuel cycle. This additional quality control needs to be carefully balanced to provide the maximum added value to the other characterization programmes, assuming the financial impacts remain reasonable.

## 2.2. TYPES OF WASTE

### 2.2.1. Definitions of new waste and historical waste

#### *New waste*

For the purposes of this publication, “new wastes” are those that are generated with a traceable characterization programme in place. They may actually have been generated some time ago, appropriately treated (not yet conditioned), and placed in interim storage; however, if the characterization was robust, traceable, necessary and sufficient, these types of wastes may be considered under the heading of new wastes for the purposes of this discussion. The primary distinction for this type of waste is that the history is known, the characterization is robust, and the process knowledge is sufficiently preserved through appropriate managerial control and custody of the wastes.

#### *Historical waste*

In contrast to the above definition of new wastes, for the purposes of this discussion “historical wastes” are those that are generated without a complete traceable characterization programme or quality management system in place. These wastes may, in fact, be generated today. For example, the continued collection and placement in storage of raw wastes without robust characterization and segregation, and which is pending retrieval and full characterization/treatment, etc. at some time in the future, is actually continued generation of historical wastes.

Key characteristics of historical wastes are:

- may be conditioned, partially treated, or raw;
- poor or no information/traceability;
- cannot conclusively identify originating process/location; and,
- waste streams may be mixed.

The primary identifiers of historical wastes are:

- incomplete history;
- incomplete or improper characterization/treatment; and
- quality system does not cover the whole life cycle at the time of generation or does not meet the modern standards for the whole life cycle.

### 2.2.2. Waste subtypes

Waste streams from both new and historical waste types may conveniently fall into one of four subtypes, which are developed as a combination of two components depending upon the consistency and complexity of the waste properties (physical, radiological, chemical, etc.). The first component is due to the complexity of developing the nuclide vector or collection of properties, and the second component is due to the variability of vectors.

The first component is described as either *simple* or *complex*. This refers to the level of difficulty (and corresponding expense) to develop the list of properties. For example, simple waste streams do not require much effort or expense to develop the vector. More complex streams will require a great deal of expensive analysis to develop the fingerprint.

The variability component refers to whether the properties remain relatively constant over time. For example, streams where the waste always comes from a consistent process will be stable, and wastes coming from varying or multiple processes are likely to be variable. Stable streams will tend to have a lower uncertainty in the vector. Variable streams will tend to be difficult to manage, as the uncertainty in the vector will vary and could be quite large. If the degree of variation is unacceptable, then it may be necessary (if possible) to revisit the stream and partition it into more stable streams.

The above two components can be further described as four subtypes, which are further described in the following paragraphs and summarised in Table I.

TABLE I. CLASSIFICATION OF WASTE STREAMS BY EASE OF MEASUREMENT AND SAMPLING ABILITY

<b>Traceable Waste Stream:</b>	Simple & Stable	Complex & Stable	Simple & Variable	Complex & Variable
Nuclear Power Plant	✓	✓		
Institutional	✓			
Nuclear Research Lab			✓	
Reprocessing		✓		
Enrichment, Conversion, Fuel Fabrication	✓			
Decommissioning	*	*	*	*
Spent Sealed Source	✓			
Spent Fuel	✓			
Final Waste Form		✓		
<b>Non-Traceable Waste Stream:</b>				
Historical	**	**	**	**

\* facility dependent.

\*\* may or may not include this subtype.

*Simple & Stable*

These waste streams are the ideal case from a characterization point of view. It is often desirable when considering new wastes to design the process, or characterization programme to maximise the waste streams of this type, as opposed to the other, more complex cases. Waste streams of this type follow consistent patterns of properties that do not vary considerably and are highly predictable. In addition to being uniform, there is not a broad

range of complex individual properties requiring consideration (e.g. complex chemical mixes, complicated radionuclide vector, etc.). As the properties do not vary, representative sampling is relatively easy, and simple measurement methods that may only capture one key property, coupled with process knowledge confirming that other properties are not varying (i.e., quality control regimes), may be sufficient to characterise the stream. For example, for wastes that contain a single gamma emitting nuclide, dose rate or gross gamma measurements are usually sufficient to characterise the radioactive properties. Examples of this subtype include the following:

— Spent sources

Constituents will be well known, and detailed documentation will be available. Sampling will not be necessary, and direct quantification will likely be possible by very simple means.

— Enrichment, conversion and fuel fabrication

The radioactive species will only be the fissile material. The nuclide vector will be known, as this is a highly controlled part of the process. The significant process control effort required for manufacture will be a valuable source of waste characterization information. As the process is highly controlled, the streams will be stable for each particular batch. (Note: It is recommended to clean out process lines between batches to preserve the stability of the waste streams.)

— Institutional and radio-pharmaceutical wastes

Similar to fuel manufacture, these processes tend to be highly controlled, with very few species. Process control data will be highly valuable to characterise wastes. Wastes will be stable within batches.

— Spent fuel

Spent fuel will not change in composition until it is reprocessed. Although nuclide ratios will change due to decay, this is a very well understood process and can be predicted very accurately. Burn-up calculations may possibly generate much of the data required to determine the radioactivity characterization.

*Complex & stable*

Some waste streams are necessarily complex as they result from complicated chemical or nuclear processes. Many properties require consideration, but if these streams are predictable (although complex), the property distributions do not vary significantly; therefore they may be considered as being of this subtype. This is the second most desirable situation for new wastes. The complex nature of the wastes means that more complex measurement approaches may be required. For example, wastes with a complex radionuclide vector may require gamma spectroscopy measurements to characterise the radioactive properties. However, as the properties are broadly non-varying, representative sampling is relatively easy. Examples of this subtype include the following:

— Nuclear power plant (NPP) operational wastes

The majority of NPP operational wastes will be highly system-correlated for example, systems associated with heat transfer will have mixed fission and activation products, while reactor components will have mainly activation products. The distributions will be highly chemistry-correlated. Other than where chemical processes are happening on the wastes, the streams will be relatively stable. Although these streams will have relatively complex nuclide vectors, the streams should be quite strongly process system/geographically-correlated and should be easy to segregate into stable streams. For example, outage wastes should be kept segregated from general operational wastes, from laboratory wastes, from heat transport/fuelling maintenance wastes, etc. Examples are protective clothing, filters, resins, decontamination wastes, etc.

— Reprocessing

Reprocessing involves complex chemical processes on spent fuel. The chemistry and dissolution processes will generate complex products, and therefore complex vector wastes. However, these are highly controlled processes; so batch to batch, the product and associated wastes will be very stable and will relate directly back to the characteristics of the particular fuel being reprocessed. There will be multiple streams, as parts of the process associated with fissile material recovery will be very high in alpha activity, and processes involved with the high level liquid waste will contain high concentrations of fission products and activation products.

— Waste form

After conditioning, the waste form will be complex. However the purpose of conditioning is to stabilise the wastes for long term disposal, so the properties at this point in time will be stable.

*Simple & variable (e.g. laboratory)*

Some wastes may not possess a wide degree of complex properties, but the properties that they do possess are highly variable. For these wastes, measurements are performed relatively easily due to the normally simple distribution of properties. However, if the properties vary considerably, representative sampling becomes quite difficult, as it is difficult to defend an assumption that a single sample result is representative of the mean of the property for the waste population. Many more measurements are required; sometimes for each waste item.

— Operational laboratory wastes often fall into this waste subtype. (Quality checking laboratories will more appropriately fall under Simple & Stable subtype.)

As laboratories perform many differing analyses, all with different reagents and source materials, the properties of the wastes will be quite variable. However, as each analysis will be highly controlled, the wastes from each individual operation will be relatively simple.

### *Complex & variable*

The least desirable situation is one for which the wastes possess a complex mix of properties, some or all of which display a large degree of variation. For this situation, representative sampling is very difficult, and measurement methods will be necessarily complex. It is not difficult to recognise that this situation usually leads to the most costly characterization programmes. Examples of this subtype of waste include decommissioning wastes and legacy stored wastes where history has been lost and streams have been mixed. In each of these cases, a large degree of costly complex sampling and analysis will be required as a foundation to the characterization programme. It should be noted that decommissioning wastes and historical wastes are unusual cases that, depending on the circumstances, could fall under any one of the four sub-categories. As such, they are given separate treatment in Section 4.

### 2.3. REQUIREMENTS

Many of the objectives of waste characterization are described in reference [2]. If the waste package is to be accepted for disposal, performance in its final form needs to be specified and assured. LILW and HLW must be characterized at various stages in predisposal management to obtain information on its properties. This information can be used in controlling the quality of the products, verifying the processes and facilitating the subsequent steps for safely processing and disposing of the waste [3–4]. To ensure that the waste package is in an acceptable form for disposal, a programme should be established to develop a process for conditioning that is approved by the regulatory body. Features adopted for waste characterization and process control should ensure that the properties of waste packages will be maintained; otherwise the long term safety demonstration of the proposed disposal facility may not be conclusive.

It is important to define what characterization details are required. With too little data, the most appropriately engineered disposal may not be selected. Too much data (and the associated data collection costs) result in a waste of resources.

Characterization must be carried out correctly. If inaccurate data is obtained, then the resulting packages may be found to be non-compliant late in the life cycle, which will be expensive to correct. If errors go undetected, the result may be unsafe disposal with potentially serious long term effects on the environment. A Quality Assurance programme should be implemented [4] as well as a system of records for keeping all the waste data [5–6].

Waste characterization requirements are typically developed from disposal performance assessment in addition to waste acceptance criteria (WAC), process control and quality assurance requirements, transportation requirements, and worker safety requirements. A matrix showing where each WAC originates can greatly assist with understanding the philosophy behind the overall characterization programme and put the elements into context.

### 2.4. LIFE CYCLE

Radioactive wastes typically undergo the following stages:

- Planning (development of characterization process)
- Waste generation or retrieval

- Treatment
- Conditioning
- Interim storage
- Disposal (including closure and post-closure of disposal facility)

Activities that are performed in various stages of the life cycle may have significant effects on the cost and efficiency of the overall characterization programme. Characterization is generally much easier and cheaper in the earlier stages of the life cycle. For example, waste properties that could easily be measured at the raw waste state may be difficult or impossible to measure after some treatment stages, certainly after conditioning has been undertaken. If wastes streams are appropriately segregated and controlled early in the life cycle, then a greater proportion of the wastes may fall into the *simple & stable* waste type. Alternatively, if raw waste streams are mixed and if valuable history is lost, more of the waste will fall into the *complex & variable* type, requiring a characterization programme that is more intensive and costly.

#### **2.4.1. Planning and waste generation**

Characterization is the assessment of key parameters, some of which include the following:

- waste volume/quantity
- activity
- nuclide vector
- chemical properties
- physical/mechanical properties
- biological properties
- originating process/location

The planning stage identifies the characterization requirements for the remaining life cycle phases. Planning for waste characterization should take place in advance of waste generation to ensure that waste collection, segregation, containerization, temporary storage, etc. are all accomplished in a controlled manner which is best supports the waste characterization plan. In general, the better the planning, the more cost effective and successful the characterization programme.

##### *Planning for new waste generation*

For new wastes, there is the opportunity to look at the originating process and influence it to avoid potential pitfalls. Analysis of waste streams followed by development of suitable segregation techniques usually result in wastes that are easier to characterize and manage. Robust institutional control of the wastes during later stages can be established and built into the system to preserve process knowledge. As a result, complex analysis or measurements beyond the waste generation phase may not be required other than the minimum required for conditioning process control or demonstration of quality assurance (e.g. independent verification). Requirements for characterization for each waste stream during each phase of the life cycle will be identified, and appropriate analysis of the engineering processes will be undertaken to design the optimum characterization programme.

## *Planning for historic wastes*

Considering that historic wastes have already been generated, there is little or no opportunity to influence the generation process. The first stage of planning will concentrate on identifying what is already known about the wastes and what characterization is required for further phases. This process is called scoping. Piecing together the strongest possible process knowledge information from historical sources (records, reports, interviews with past employees, etc.) may help reduce the amount of expensive sampling and analysis required to complete the picture. Scoping studies will identify the boundaries between properties that result in logical waste streams being identified. The characterization programme will then be developed around gaps in knowledge until all the waste streams have a sufficiently robust characterization.

### **2.4.2. Treatment**

Some wastes may require treatment for safety, handling or stability for interim storage reasons. For new wastes, there is an opportunity to influence the process design so that wastes generated will require little or no treatment. If treatment is required, it is usually easier to obtain most of the characterization while the waste is in raw form, and characterization requirements may be directed toward treatment process control.

For historic wastes, many situations are possible. Wastes may have already undergone some degree of treatment with little or no pre-characterization. In such a case, further characterization will be required both before and during treatment to obtain a sufficient degree of detailed information. Waste streams may have been inadvertently combined, leading to a much larger volume of material that must be checked for certain properties. Previously treated wastes need to be examined to determine the compatibility of the prior treatment process with the waste acceptance criteria for the conditioning and disposal phases.

### **2.4.3. Conditioning**

By the time this phase occurs, sufficient characterization should have been accomplished to comply with the WAC for conditioning and disposal, and the waste will likely be in an acceptable form. During this phase, characterization activities are primarily directed toward quality control, although a minimum amount of independent quality checking may also be required. cursory checks will be directed toward confirming the identity of the incoming wastes. Process knowledge and characterization done in previous steps, combined with knowledge of what impact the conditioning process has had on the waste characteristics, will form the basis for overall characterization of the waste and waste forms. The growing set of documentation for each waste package preserves the history and characterization throughout the remainder of the waste life cycle. Post-conditioning checks will confirm compliance with requirements for transport (e.g. surface contamination levels, dose rate, etc.).

### **2.4.4. Storage and disposal**

At this stage, there should no longer be any requirement to sample or analyze waste forms, except to verify randomly that waste delivered to the facility complies with the waste acceptance criteria. Suitable management control throughout the preceding phases, coupled with robust quality assurance and quality control, will make re-analysis unnecessary. Any destructive analysis at this stage would pose considerable technical difficulties, be unlikely to

give a complete picture, and would be very expensive. All that should be required during this phase is compiling basic information for the protection of the workers handling the wastes (e.g. dose rate, surface contamination levels).

## 2.5. DEVELOPMENT OF CHARACTERIZATION PROGRAMME STRATEGY

In order to establish a characterization programme, it is necessary to determine all of the requirements.

The easiest case occurs in a jurisdiction where each phase of the life cycle is in operation and the regulatory regime is well developed. In other words, the industry local nuclear is mature and final waste disposal is actually occurring. Given that the disposal concept will influence the conditioning and storage phases, it is best to start from the requirements (or WAC) [11] for disposal and work back from there.

If no mature disposal concept has been established, one has to work forward and try to presuppose what the disposal future requirements will be. This situation inherently contains much more risk, given that overlooking parameters may have severe monetary consequences. It will also be difficult to predict what level of analysis will be required to satisfy the regulator.

Developing a programme for this second and more difficult case will require a measured approach. On one hand, it may seem advantageous to defer characterization activities until the requirements are more clearly known. However, the risk of having to gain necessary information late in the life cycle and incurring high costs to do so could be great. Extensive testing may seem desirable to avoid this risk, but it would seem unnecessarily costly if expensive tests are performed and shown later to be unnecessary. Archiving of samples is a good way to help reduce this risk, considering that the samples will be available for future testing. However, this is also very complicated because choosing which samples would be valuable in the future is difficult to predict. Consideration must also be given to preserving the long term stability of the samples as well as managing and storing the samples.

A practical place to start is with consulting the stakeholders involved, especially the regulators, and studying the approaches taken in jurisdictions that are more mature. At an absolute minimum, the requirements of packaging and interim storage must be considered if there is not yet a disposal concept, and it would be wise to consider the possibility of interim storage being necessary for a very long time. It would be better to develop a conditioning-for-storage strategy that is consistent with the principles for disposal and determine the characterization requirements for the generation, packaging, transport, treatment and conditioning processes.

### 2.5.1. Principle issues and development of waste acceptance criteria

As previously stated, the characterization programme must necessarily address the requirements for all life cycle phases. This may involve requirements from several different waste acceptance criteria publications and additional regulatory or political requirements. It is important to note that this picture will have to be generated independently for all waste types, and not all wastes will be destined for the same processes and disposal concept.

The waste acceptance criteria are likely to contain a mix of requirements, some of which pertain to key characterization parameters. Others may be related to documentation requirements, contractual constraints, limitations of the waste receivers processes or equipment (e.g. the waste receiver may have limitations on crane size and cannot accept packages over a certain weight). Whoever is responsible for developing the characterization strategy, it is important to study and interpret any relevant WAC to understand fully the reasons behind each requirement. This will highlight which requirements may be negotiable or may require further explanation and allows for optimal tailoring of the characterization process.

If the waste management regime is not fully mature and detailed WAC do not exist for the entire waste life cycle, it may be necessary to presuppose what these requirements are likely to be as the waste management programme evolves. In order to do so, it is helpful to review more mature programmes and analyze in detail the potential requirements for each phase of the life cycle. Of course, all of this information will have to be adapted to the organizational, political, economic and regulatory constraints of the local jurisdiction.

#### *Elements of WAC and where they originate*

The waste acceptance criteria will define the requirements that the waste package must meet in view of transport, interim storage and final disposal. The requirements will be based on the following:

- limitations of conditioning processes or facilities
- conditioning process control parameters
- worker safety at all phases
- legal requirements
- transportation limits
- interim storage requirements
- integrated performance assessments
- disposal facility performance assessments
- overall quality assurance requirements (independence, testing, etc.)

#### **2.5.2. Elements of characterization programmes**

Waste characterization involves a collection of data that pertains to specific waste properties as well as processing parameters and quality assurance. Key elements include the following:

- Radioactivity properties
- Chemical properties
- Physical properties
- Mechanical properties
- Thermal properties
- Biological properties

The following parameters relate to individual waste packages and must be known to ensure that each individual package is managed correctly:

- Waste stream identification

- Description of originating process
- Date and location of waste collection
- Names of organizations and persons involved at each step
- References to exact procedures used
- Traceability of reference materials to consensus standards
- Training and qualifications of persons involved
- Dates and times of analysis
- Acceptance criteria for quality control tests
- Results of quality control tests
- Required frequency of quality control tests

This information serves three principal purposes. The first is to provide additional backup information to describe the wastes. The second purpose is to support quality control and ensure that the waste processes (e.g. conditioning, packaging) are kept within tolerance limits to ensure compliance with relevant WAC. The third purpose is for quality assurance. This data confirms that everything continues to be carried out correctly and provides confidence to the stakeholders. Whenever a non-conformance is detected, it is crucial to determine where the breakdown occurred so that the problem may be corrected.

### **2.5.3. Quality assurance and quality control**

A quality assurance programme for predisposal management must be established and implemented by the operator of the facility concerned in accordance with the requirements and recommendations for quality assurance provided in Reference [7]. Non-conformities in waste packages should be prevented, particularly for those activities that could lead to an irreversible non-conformance if not performed properly. A quality assurance programme, including implementing procedures, should involve the following activities:

- (a) Characterization of the waste;
- (b) Development of the specifications for waste packages ;
- (c) Approval of the conditioning process for the waste;
- (d) Confirmation of the characteristics for waste packages; and
- (e) Review of quality control records.

Quality records should be established and maintained for each waste package. These records should be reviewed against the specifications to determine the acceptability of the waste package. Records generated at all predisposal stages may be important for demonstrating that the waste package complies with the specifications. Such records should ensure that characteristics of the waste from its collection through to its processing and storage are known, documented and retrievable [5, 6, 13].

## **2.6. ORGANIZATION AND RESPONSIBILITY**

Figure 2.1 shows the responsibilities of various players involved in a waste characterization programme [3, 4].

The regulator oversees every step in the waste characterization programme. The producer of raw waste has to conduct characterization of the waste he produces. The treatment

operator must conduct quality control during the process. Finally, the disposal operator must see that the final waste package conforms to criteria for disposal.

Taking into account the particular activities involved in each step and in the whole life cycle can help fulfil all requirements in a more effective way. Each body has to obtain regulatory agreement/approval as well as agreement from the next body (operator or authority) to transfer the waste to its next life cycle phase. It is crucial that information about the waste (including its characterization results) from the generator to disposal operator is traceable and shared between bodies.

### **2.6.1. Responsibilities of the regulator/licensing authority**

The regulator (licensing authority) has related responsibilities which begin prior to operation of a waste characterization facility and continue through final closure. These include:

- Review of the Safety Analysis Report (SAR) and granting of the license.
- Issuing license requirements, including waste acceptance criteria (or at least activity limits)
- Quality audit
- Independent verification of characterization accuracy and reliability
- Radiological safety inspections
- Technical inspections
- License amendments and extensions, including review of SAR revisions
- Characterization facility (e.g. laboratory) decommissioning and license termination

### **2.6.2. Responsibilities of the generator of the waste**

The waste generator has ultimate responsibility for characterizing the wastes so that the Waste Acceptance Criteria can be met throughout the life cycle. In principle, the generator should be prepared to cover all costs of waste management including waste characterization. The generator can design the whole life cycle, identify each waste transfer between various bodies, and collect the criteria and requirements that must be fulfilled when waste goes through each phase. In most cases, effective characterization can be done on the raw waste. To be able to use early life cycle waste characterization results in subsequent phases, the generator should set up an extensive quality control system with special attention given to preserving obtained data and its proper management in subsequent waste management steps. Ideally, all players in this process will manage data in an equally effective and secure manner. However, if the subsequent operator cannot handle his publication management system in a way that covers all needs during the waste management life cycle, it is ultimately the waste generator's duty to collect and properly handle data until it is accepted by the disposal operating company.

The generator of the waste is responsible for the quality control of the waste. The quality control system shall include training, method development, qualification and documentation. Preserving the characterization data for traceability and retrievability is another important responsibility for the generator.

### **2.6.3. Responsibilities of the treatment/conditioning/storage operator**

Before treating and conditioning the waste, the conditioning operator has to qualify treatment and conditioning methods as well as methods for characterizing the process an end product. This activity has to be licensed by the regulator.

When the waste arrives at the conditioner, he has to inspect it and perform quality control checks to be sure the waste fulfils the waste acceptance criteria.

Quality control also has to be done during the treatment/conditioning process. If the waste packages will be stored after conditioning, they must be inspected to ensure that nothing has happened to the waste packages while under his control.

Continued preservation of the characterization data for traceability is another important consideration.

If secondary waste arises during the treatment/conditioning process, the operator is responsible for characterizing that waste too.

### **2.6.4. Responsibilities of the operator of the disposal facility (operating company, agency)**

The operating company/agency is responsible for licensing the repository and operating it in accordance with license requirements. This includes performance assessment, quality system set up, safety analysis, and developing the waste acceptance criteria. This includes defining the waste acceptance criteria consistent with the operating license and regulatory requirements, and it includes verifying compliance with the waste acceptance criteria.

When the waste arrives at the repository, the operator must inspect the waste and check the enclosed publications to be sure that the waste fulfils the waste acceptance criteria.

It may well be that not every item of the waste acceptance criteria can be readily tested upon receipt (or tests may be very expensive). In those cases, the operator's responsibility is to inspect, check and document those items that cannot be inspected during waste acceptance procedures at the repository site.

It is very important to keep all documentation of the disposed waste packages; the operating company/agency is responsible for carrying out this activity. Some requirements may be changed during the waste life cycle, and only proper documentation covered by a quality system enables later checking.

The repository operator/agency is also responsible for the post closure monitoring if necessary.

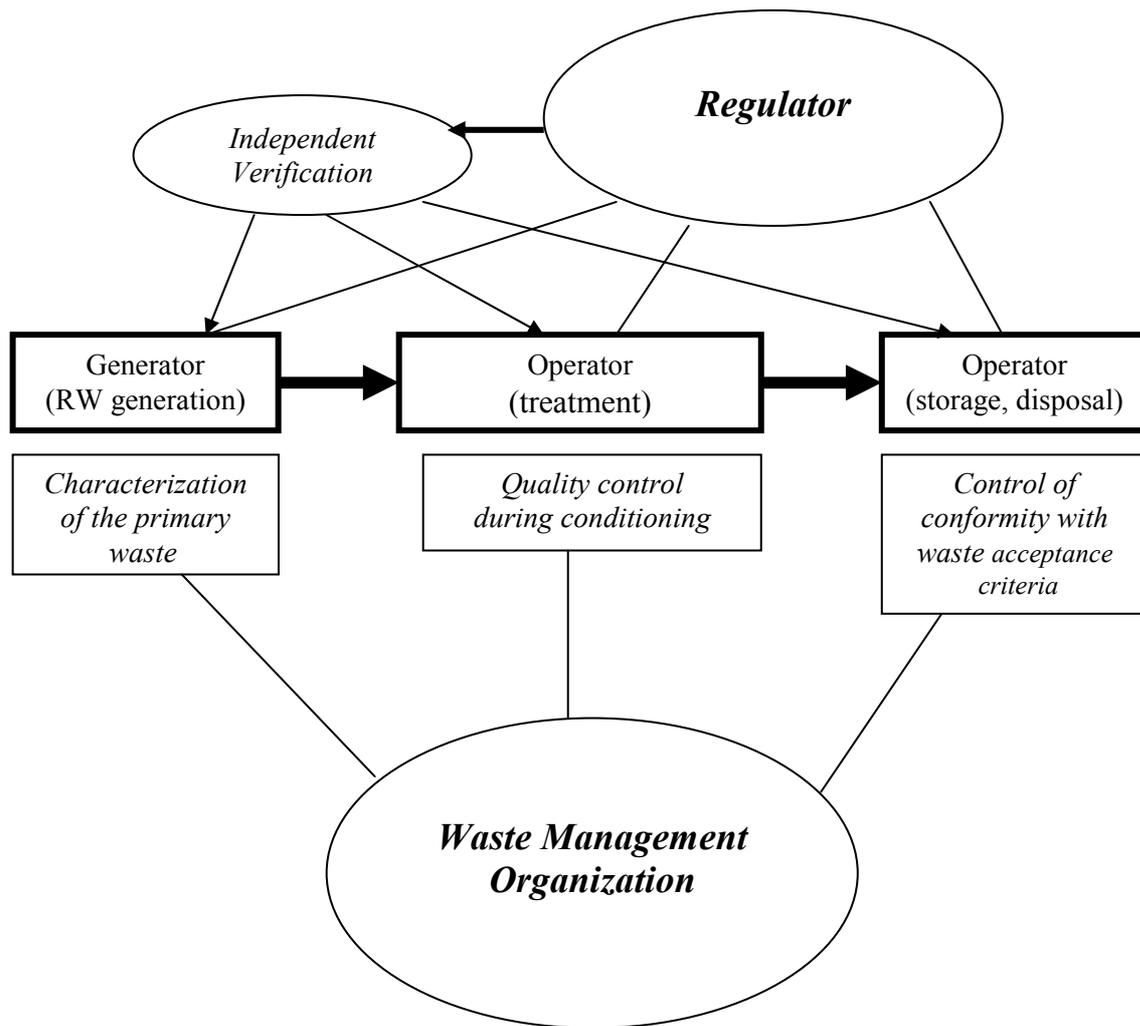


FIG. 2.1. Diagram showing responsibilities of various players in a waste characterization programme.

### 2.6.5. Independent verification

Independent verification is a crucial element of the overall waste characterization system, especially when responsibility for waste characterization is distributed between several bodies or organizations. Intensity (frequency) of independent verification depends on the phase of the life cycle and the particular regulatory and organizational regime. Generally, the early steps in the process need more frequent and intensive verification than the last step (disposal). Evidence of proper waste handling and characterization is necessary for all operators.

All phases during the waste life cycle are licensed and supervised by the regulator or authority, and independent verification helps to show regulator independence. The degree of independence that the testing body (laboratory) has will vary. There are practical problems due to the limited number of bodies skilled enough to carry out independent verification and

prove independence from the operator. This can possibly be solved by implementing international cooperation.

## 2.7. COST AND BENEFITS OF WASTE CHARACTERIZATION

Necessary and sufficient criteria for waste characterization must be identified as early as possible in the whole life cycle of waste management. The greatest benefits of waste characterization occur when the main investments and operating costs of characterization efforts are undertaken by the waste generator as early as possible rather than at later stages of the waste management life cycle.

Waste characterization uses three main techniques:

- Process knowledge
- Non-destructive examination and assay (NDE and NDA)
- Destructive analysis (DA)

### 2.7.1. Process knowledge

Knowledge and documentation of the processes giving rise to waste generation is one of the most effective ways of assessing the characteristics of waste. Process knowledge can help limit the range of characteristics possible and analysis methods to be used. In a fuel manufacturing plant, for example, uranium and its attendant products may be the only radionuclide species possible; therefore, broad analyzes to identify components in the nuclide vector will not be necessary, and specific methods to quantify the uranium present will be all that is required. For new wastes, maximum benefit of process knowledge is being obtained by setting up effective waste stream control and segregation practices at the point of origin.. A majority of the waste streams may fall into the simple and stable subcategories.

Process knowledge can also be valuable when considering historical wastes. Say a waste drum is discovered in a corner of the plant and a label on the drum indicates where the drum had previously been e.g. Fuel handling. Then say a similar drum is found in a waste storage area with no labelling or documentation.

In the first scenario, there is some process knowledge, i.e., the drum was at one time located in the fuel handling area of the plant. Given that fuel machine maintenance often uses organic solvents for degreasing, the waste manager is likely to plan a waste characterization strategy assuming the drum may contain an organic liquid containing radionuclides present in fuel. The plan of course will need to validate the assumption but once confirmed the waste can be more easily and cost effectively characterized for both conventional and radiological contaminants.

In the second scenario, the waste manager has a drum of waste but no information on its contents. As a result, the waste characterization strategy will be very broad and will require many assumptions to be validated by NDA and DA, possibly at great expense.

The lesson for waste generators from these examples is this: Provide suitable process knowledge documentation, and ensure it is linked to the waste container either by labelling or barcoding. Even if the waste container contents are not completely characterized, any amount

of documentation will provide a road map by which waste managers can establish a cost effective waste characterization strategy.

### **2.7.2. Timing of waste characterization efforts**

Costs associated with documenting and disseminating process knowledge are relatively low. However is not true for the other techniques for waste characterization, namely, NDE, NDA or DA. The technology available today for NDE and NDA is reliable but costly. It is therefore quite reasonable for controllers to ask whether such equipment and characterization costs can be deferred. If waste is simply being stored and not finally disposed, it is reasonable to assume that the characterization for waste disposal would have a lower priority than in a jurisdiction that practices waste disposal. So what should the waste manager do: Characterize the waste early in its life cycle? Or defer characterizing waste until it is ready to be disposed of?

The nuclear industry has found the following:

- When characterization is conducted at the point of waste generation or production, it can be carried out more simply and accurately without having to make assumptions that need to be validated.
- Characterization will be less expensive when performed at the earliest practical stage in the waste life cycle. Early waste characterization also has safety benefits: the waste is likely to be handled less, samples are likely to be representative, and waste characteristics are likely to be more accurate.
- Incomplete waste characterization often leads to reliance on overly conservative assumptions, which are costly. Early waste characterization provides greater accuracy for determining waste characteristics and, thereby, better utilizes waste storage and disposal facilities.

It is also the view of the radioactive waste management community that the more waste generators know about their waste, the more convincing they can be in front of the regulator or community regarding waste management strategies. This will lead to greater public confidence.

It is also evident that there needs to be a financial incentive or benefit for the waste generators to characterize their waste to the extent that is sufficient and necessary for safe waste disposal at the earliest possible moment in the life cycle.

## **3. IMPORTANT FACTORS TO CONSIDER IN DEFINING A WASTE CHARACTERIZATION PLAN**

### **3.1. SCALING FACTOR METHODOLOGY**

The inventory of the radionuclides to be declared with each waste form is numerous and varied (alpha and/or beta emitters). Some of these radionuclides are easy to measure (ETM) using NDA (non-destructive assay) or DA (destructive assay), but most of them are difficult to

measure (DTM) and need DA in a laboratory or a calculation using special codes (e.g. pure beta emitters). Some of them are impossible to measure (ITM) even in a laboratory.

The scaling factor methodology [1] can determine the radioactivity of DTM and ITM radionuclides using correlations between them and key nuclides (KN) chosen among the ETM. Specifically, the DTM nuclides are predicted from a gamma nuclide easily measured by multiplying the concentration of this KN by the scaling factors calculated from the radioactivity of nuclides obtained through appropriate radiochemical analysis or through modeling code calculation, and which represent the average relationship of the DTM nuclide to the KN. The ITM nuclides are also predicted from an ETM gamma nuclide, best is done by calculation using a model such as ORIGEN or MCNP.

Each waste package can be measured with an NDA system, which provides the KN concentrations. By using the scaling factors, the associated DTM, ITM and ETM nuclides are then calculated and declared for each package. When NDA measurements provide the KN and other ETM nuclides, these data are kept for periodic control of the validity of the scaling factors used for the inventory. This methodology shows good applicability for stable waste streams. It is also applicable for simple and variable waste streams with higher correlation uncertainty. Its applicability for complex and variable waste streams is difficult. Scaling factor methodology requires sampling, destructive analysis, modelling, non- destructive analysis, and calculation.

## 3.2. ACCURACY CONSIDERATIONS

### 3.2.1. Introduction

The purpose of this section is to identify the main sources of uncertainty in the waste characterization process. It does not put forth rules of good practice for estimating the radionuclides and non-radioactive elements in a particular waste stream; it instead highlights the true objectives and the main sources of error.

If the long term safety of a repository depends on total or mean values of the radioactivity or toxic elements of the disposed conditioned wastes, the level of confidence and the accuracy gained for the composition of a single package should be of minor importance. In other words, the primary requirement of the methods used to declare the radionuclides and environmentally important non-radioactive elements is that they must be free from any significant systematic errors. In this case, accurate results can be obtained for the entire lot using rapid and inexpensive measurements of each container.

The main sources of uncertainty arise in checking the homogeneity of a waste stream and sampling, selecting the key nuclides and non-radioactive elements, measuring the easy and difficult to measure radionuclides (by using NDA, DA including the dissolution step), and calculating the impossible to measure radionuclides.

Waste characterization samples will range from liquids to slurries and solids to final waste forms, such as waste glass or drummed waste. The type of waste sample, depending upon the consistency and complexity of the waste properties, will influence the methods of sampling. Another important consideration that will affect sampling will be the occupational dose uptake to plant operators and laboratory personnel, which must satisfy ALARA principles. Sampling events, quantities and amount of analytes measured will also depend on

the data need and usage, e.g. for primary waste flux characterization, process control, process monitoring, contractual compliance, environmental compliance, safety basis compliance, technical investigations, and commissioning/decommissioning.

For these reasons, an integrated sampling and analysis publication should be developed early in the process and agreed upon in conjunction with the waste generator, treatment and conditioning plant owner and operators, storage and disposal personnel, and regulators. Waste compliance and regulatory requirements would be included in this publication. Determining the time that samples will be gathered and analyzed, including all quality assurance and control data, is important because it will have a direct impact on the throughput of the treatment or conditioning process. Based upon negotiations between participants, sampling and analysis by an independent laboratory may also need to be included in the plan. This type of development approach leads to increased communication and agreement between waste generators, flowsheet developers, plant and laboratory designers, owner/operators, and regulatory and governmental agencies.

### **3.2.2. Checking the homogeneity of a waste stream and sampling**

#### *3.2.2.1. Homogeneity of a waste stream*

Before sampling, the homogeneity of the distribution of radioactivity in the waste stream has to be checked. When possible, this should be done, on the primary waste flux. The homogeneity is obviously easier to demonstrate for a stable waste stream than for a variable stream. It is necessary to define which radionuclides, non-radioactive elements and other properties will be measured, how they will be measured (NDA and/or DA measurements), and what the criteria will be to facilitate the development and design of appropriate treatment and conditioning processes.

For stable waste streams, measuring one or more key nuclides and non-radioactive elements may be sufficient to check the homogeneity. For example, a simple and stable waste stream could be declared homogeneous if NDA measurements of  $^{137}\text{Cs}$  and/or  $^{60}\text{Co}$  made at different locations are within a 30% relative interval. The number of samples required to provide radionuclide scaling factors will depend on this value and the acceptable uncertainty negotiated with the regulator.

For variable waste streams, the difficulty in generating a sufficiently representative sample will increase greatly, especially as one moves toward very complex waste stream mixtures. For example, sufficient mixing energy will need to be introduced to a vessel to mix the waste contents so that representative samples can be obtained. A method to demonstrate representative slurry samples from a vessel is given in Section 4.7.1.2. Methods to generate sampling and analysis plans of nuclear materials and contaminated soils are provided in Annex III, Relevant Standards.

Finally, it is critical that representative primary waste flux samples are taken over the range of wastes expected to be conditioned and to characterize them sufficiently. By examining chemical, physical, and rheological properties, the required processing limits may be defined. The initial and subsequent characterization results will be used to generate accurate testing simulants (see Sections 5.4.2 and 5.4.3) and will aid in designing the treatment and conditioning processes.

### 3.2.2.2. *Sampling*

Based upon the complexity of the waste, the number of operations required to condition the waste stream, and the characteristics of the final waste form, a process control and monitoring scheme must be determined [20]. For a stable waste stream with simple conditioning operations and a consistent final waste form, sampling and analysis of the final waste form may be sufficient for process control and monitoring, i.e., a feedback control strategy. For a complex, variable waste stream requiring multiple integrated operations to condition the waste and process it to a final waste form, a number of sampling points may be necessary to provide sufficient control and monitoring of the conditioning process, i.e., a feed forward control strategy. A feed forward control strategy will be valuable for final waste forms such as canistered high-level waste glass, which is difficult to recondition if found to be outside of an acceptable composition range. The conditioning process control strategy, sampling points, number of samples and analyses completed should be discussed and agreed upon with all participants and documented in an integrated sampling and analysis requirements publication. This sampling plan will need to detail the necessary quality assurance (QA) and quality control (QC) requirements and analytical procedures. The sampling plan and subsequent analytical documentation should ensure that the procedures meet all applicable levels of accuracy, reliability and precision needed to control and monitor the conditioning process adequately.

As mentioned earlier, the characterization results of the waste stream may be used to develop simulants for subsequent designing, testing, and building waste conditioning facilities. Testing and sampling needs should be conducted on a suitable scale with representative equipment to demonstrate that the conditioning process and the sampling system work. Sampling methods may include manual methods such as grab samples; automated sampling such as taking a sample from a recirculation line and remotely transporting it to the analytical laboratory; and inline instrumentation rather than laboratory analysis. Typically, the sampling process is automated for high frequency and high radiation samples. The process must be able to mix the waste representatively to meet the conditioning process requirements, to obtain representative samples at the sample point (vessel or pipe), and to transfer representatively both the sample and waste to the next unit operation [21]. The chemical and radiochemical composition variability of the mixing, sampling, and transfer systems will need to be tested and quantified over the range of maximum and minimum chemical, physical, and rheological parameters [22].

Recommendations include the following:

1. An integrated sampling and analysis requirements publication should be developed early in the process and agreed upon in conjunction with all participants, e.g. waste generator, waste conditioner, independent laboratories and regulatory agencies.
  - a. Occupational dose uptake to plant operators and laboratory personnel will need to be considered.
  - b. The time to gather and analyze a sample, including all quality assurance and control aspects, must be factored into the conditioning process.
  - c. The conditioning process control strategy, sampling points, number of samples, analyzes completed, etc. should be discussed and agreed upon. For example, defining the number of samples depends on the acceptable accuracy, which may be subject to negotiation.

2. Sampling and analysis of the primary waste stream will provide the most accurate characterization data.
  - a. Sampling of the primary waste stream must demonstrate that it is representative with minimal random error and statistically relevant.
  - b. Representative primary waste flux samples must be taken over the range of wastes expected to be conditioned.
  - c. A complex waste stream conditioning process will need to demonstrate the ability to mix the waste representatively, obtain representative samples at the sample point, and transfer representatively. This is especially important, given that most currently stored waste is heterogeneous.

### **3.2.3. Selection of the key nuclides**

To establish representative correlations, KN are selected according to the following criteria:

- The KN must have a correlation with DTM and ITM nuclides;
- The KN must be present in the assay mixture in significant quantities;
- The radioactive half-life of the KN must be long compared with the time necessary for sampling, measurement, and physicochemical processing;
- The KN, DTM and ITM in the initial mixture should be formed by a similar mechanism: in nuclear fuel cycle facilities, the radionuclides arising from fission reactions in the fuel should be distinguished from radionuclides due to activation of structural materials;
- The chemical behaviour (e.g. solubility) of the KN, DTM and ITM in the process steps should be similar; it is preferable to choose isotopes of the same chemical element; and
- The KN can be non-destructively measured beyond the detection limit.

### **3.2.4. Measurement methods**

The quality control and quality assurance of different waste streams and waste forms require that the nuclide inventory, as well as other physical and chemical properties, be determined for their production, conditioning, transport, interim storage, and final disposal.

International, national and state regulations determine the inventory and the parameters to be characterized for any facility and activity that involves radioactive waste management. Strategies in the field of radiological characterization need appropriate radio-analytical and radio-metrological support in order to determine the concentration of important nuclides within the relevant accuracy.

Measured radionuclides can be divided into two categories according to the analytical methodology to be applied: difficult to measure radionuclides (DTM), which cannot be characterized by direct measurement in the waste form, and easy to measure radionuclides (ETM), which can be determined by direct measurements.

#### *3.2.4.1. Destructive methods*

Characterization of waste by destructive methods leads to an accurate and complete determination of the radionuclide inventory. The reliability of these determinations regarding the waste flux to be characterized depends on the sample representativeness and homogeneity of the waste stream.

The concentration of DTM nuclides (alpha, pure beta and X ray and low energy gamma emitters) is usually determined by destructive assay. These methods consist basically of the following:

- Sample dissolution
- Specific chemical separation process
- Radiometry depending on chemical and radioactive properties of the nuclides

Application requires the use of hot cells or glove boxes for sampling, pretreatment and dissolution of the sample. The selection of appropriate dissolution methods in order to obtain a homogeneous sample depends on the physical and chemical nature of the waste and chemical properties of the elements to isolate (e.g. if a volatile material is to be determined, the treatment of the sample has to avoid exhausting the gases from the system). The dissolution step is probably the most important step in the destructive analysis. No universal chemical reagent is capable of dissolving all radionuclides in any solid waste category. The dissolution procedure must be qualified by checking its ability to dissolve a KN or DTM in solid waste. To this end, the use of reference materials is recommended. If not available, the laboratory can produce its own calibrated material or look for the most appropriate way to do the determination.

Dissolution methods are widely described in the literature, and there are examples of more common waste forms or primary waste such as concrete, bitumen, ashes, sludge, resins, fines and graphite (see Section 5).

Chemical separations must be designed to isolate the element to be determined from the chemical and radioactive interferences. Basic methodologies are liquid-liquid extraction, precipitation, distillation, catalytic combustion, extraction chromatography, ion exchange chromatography, and mass spectrometry.

The final step of a destructive analysis involves measuring the radioactivity concentration of the radionuclides. This process can be applied directly to the dissolved sample or after the separation process, depending on the complexity of the mixture and the resolution of the instrumental technique used for determination.

Counting systems for isolated alpha emitting nuclides are solid scintillation counters (ZnS(Ag)) or proportional counters. In the case of a mixture of alpha emitting nuclides, alpha-beta discrimination liquid scintillation counters (LSC) can be used, which give alpha spectra with low resolution. More accurate measurement can be achieved with alpha spectrometry with semiconductor detectors (~ 12 keV of resolution), which allows the determination but does not avoid the chemical separation process.

Pure beta emitting nuclides can be determined by proportional counters but the most effective method is LSC, which can determine beta emitters with  $\beta_{\max} \leq 2000$  keV with previous separation processes.

X ray and low gamma emitting nuclides can be determined by low energy gamma spectrometry with Planar-Ge detectors in complex mixtures. The low concentration of these emitters, when compared with the high-energy macro-component gamma nuclides (e.g.  $^{137}\text{Cs}$ ), can perform chemical separations prior to measurements. If no interference is found in a

complex mixture of X ray and low energy gamma emitters, direct measurement could be carried out.

High energy gamma emitting nuclides can be determined by direct measurement in an appropriate geometry by solid scintillation gamma spectrometry (low resolution). High-resolution gamma spectrometry with semiconductor detectors may be applied when a complex mixture of gamma emitters is treated before separation or if the waste has a simple composition of gamma emitters by direct measurement of the dissolved sample.

#### *3.2.4.2. Non-destructive assay*

The non-destructive assay (NDA) systems are the characterization tools used to determine the activity concentration of KN, which can be applied to the entire production of waste packages.

NDA techniques are based on the detection of a photon flux and/or neutron flux from waste package surfaces. The accuracy of the results depends on the complexity of the waste package being measured and the sophistication of the systems. These systems can range from low resolution gamma spectrometry to segmented gamma scanning (SGS) with transmission correction (TC), transmission/emission computed tomography (T/ECT) and from passive neutron counting to active/passive neutron interrogation combined with SGS. The most applied NDA techniques are summarized in Section 5.

The characterization by NDA for QC/QA of waste packages involves techniques that can be applied as a function of the complexity and variability of the primary waste and waste package structure. The scheme starts from the simplest measurement process to the most sophisticated (active/passive) processes. Selecting the appropriate NDA technique depends on several factors related to the response of the system and the type of waste to be characterized.

Determining the system response in NDA consists of establishing the calibration function for the item being characterized. Two different strategies can be adopted: calibration relative to standards and analytical calibration.

Calibration relative to standards can be performed in a simple way using a working standard selected among the waste packages to be characterized. This waste package should represent the complete production or set of packages assuming homogeneous physicochemical and radiological characteristics and it should be fully characterized before use as a calibrator [23].

The most widespread experimental calibration technique is the use of artificial standards. This technique involves the preparation of mock-ups that simulate the geometrical and physicochemical characteristics of the waste matrix. Depending on the NDA technique and the correction methodology, this calibration is quite accurate in determining radioactivity.

Calibration by analytical methods implies the simulation of all relevant parameters related to the waste package and the use of theoretical models and numerical simulations. Calibration based on MCNP (Monte Carlo N-Particle transport code) or combinations of MCNP and numerical integration simulation processes have recently been extended. Calibration techniques allow simulating the complete characteristics of the NDA system, comparing the item to be assayed with the calibrated equipment. The uncertainty associated to

this calibration method depends on the item to be measured [24]. This implies the use of sophisticated techniques or unbiased a priori information about activity and matrix contents.

Selecting a specific NDA technique will depend upon the radionuclides (gamma-X ray emitters or fissile material) and the relevant parameters of the waste package, especially the following factors:

- Type of matrix.
- Radioactivity/sources and density distribution.
- Shape and dimensions of the container and inner/outer shielding.

Adequate NDA techniques for radioactivity determination of KN and other nuclides can be defined as a function of data on the item and knowledge of the above mentioned parameters.

ETM nuclides in simple and stable waste can be determined by low resolution gamma spectrometry when the mixture of  $\gamma$ -emitters is not complex or when the nuclide vector is known (ratios between total gamma and activity of nuclides are defined). Gamma scanning with semiconductor detectors in integral mode gives accurate results as well. The determination of spontaneous fission nuclides can be performed by passive total counting, passive coincidence counting and multiplicity counting methods. These give enough accuracy because the amounts of fissile material in this type of waste are usually well known.

The NDA technique required for gamma activity determination in simple and variable waste depends on the origin of the waste. Due to the variable chemical origin of the wastes, high-resolution gamma scanning in integral mode or segmented gamma scanning (vertical) is recommended. Fissile material determination in this type of waste depends on the origin of the waste. Passive neutron counting can reach the desired accuracy level because the isotopic content is usually known in this type of waste. Active neutron counting is recommended when the isotopic composition of the fissile material has to be determined.

Complex and stable and complex and variable waste types require segmented gamma scanning for gamma nuclides determination and correction by energy transmission (mono-energetic or multi-energetic source) to determine the activity distribution in vertical, angular and radial mode and the density and effective (average) atomic number ( $Z_{\text{eff}}$ ) of the waste form. The scanning mode should be selected depending on the expected spatial distribution of the radioactivity:

- Vertical scanning: vertical heterogeneous activity distribution (assuming radial and angular homogeneity in each segment);
- Angular scanning: in combination with vertical scanning characterizes coordinates and activity of the hot spots in each vertical segment; or
- Swivelling/horizontal scanning: in combination with vertical scanning, determines the radial radioactivity distribution. This type of scanning is useful for unknown inner shielding (active or inactive) in the waste packages.

In the case of complex and variable and some cases of complex and stable waste fluxes, gamma imaging and/or emission/transmission computed tomography are also needed. Fissile material determination in both cases requires a combination of passive neutron counting, neutron interrogation and gamma scanning.

## *Semi-empirical Calculation Methods and Models*

To infer radioactivity, the simplest and fastest methods involve dose measurements. They are usually applied to simple/complex stable waste forms (homogeneous waste streams conditioned in uniform matrices), but they can be used for characterizing other types of waste (simple and variable) and samples of non-conditioned liquid waste. These methods are based on measuring contact dose rate at predefined locations of the waste form that are then used to calculate the mean dose rate. The mean dose rate ( $\dot{D}$ ) is correlated with the attenuation and/or self-attenuation factors as a function of apparent density for the pre-established gamma emitting nuclides (e.g.  $^{60}\text{Co}$ ) to calculate the total activity (A) through the following algorithm:

$$A = \frac{\dot{D}}{\sum d_i \cdot f_i}$$

where  $d_i$  (dose/activity factor) and  $f_i$  (isotopic fraction) are calculated for known or assumed waste form geometry, activity distribution, and physicochemical characteristics. Once the total activity is calculated, the activity of each KN in the inventory is obtained from the corresponding scaling factor.

If the physicochemical processes within the waste streams are well known, models can be used that evaluate the relative activity of certain radionuclides in the inventory as a function of the physical parameters of the waste streams. The results, however, may be inaccurate because of the physical parameters of the individual waste to be characterized. Since these limitations are unavoidable, models such as ORIGEN provide a rough value that indicates the range of the activity concentration determined by destructive and non-destructive measurements.

Calibration of the radio-metrology systems can be performed using simulation software. They combine random generation of radioactive events, and their interaction in the simulated environment and deterministic numerical simulation to simulate the system responses.

For those methods, it may be difficult to determine the input values with enough accuracy, given that the response is affected by uncertainty in the geometric and environment parameters to be considered. These systems are extremely useful and widely applied for non-calibrated and complex geometries that will be difficult to calibrate experimentally.

### **3.2.5. Reference materials, proficiency tests; associated traceability**

#### *3.2.5.1 Reference materials*

In addition to standard test methods and guides that promote public and worker safety and environmental health, quality assurance, and regulatory compliance within the nuclear industry, it is important to have standard reference materials that provide data on the physical and chemical properties of waste and waste forms.

Chemical and physical analysis data of nuclear materials, e.g. conditioned and non-conditioned wastes, waste forms and packages may need to withstand legal challenges to demonstrate waste material storage, transport, and disposal qualification and licensing

activities. Development and use of standard reference materials, both simulated non-radioactive and radioactive materials, allow laboratories to compare the quality of analytical methods using different techniques and instrumentation, provide a means to improve accuracy and precision of methods and techniques, allow for direct comparison of analytical results, both intra- and inter-laboratory, when used as blind samples and allow for bias correcting of analytical results when processed alongside actual waste samples.

Determining the quality of analytical data for the primary waste flux during conditioning processes and on the final waste form will allow one to make intelligent decisions on which conditioning process to use, will help taking decisions during conditioning to meet control and compliance strategies, and predicting long-term waste behaviour. The use of standard reference materials will allow operators to meet these constraints by documenting the quality and analytical reliability of the data, help in the selection of facilities and instrumentation to meet processing needs, evaluate analytical methods/procedures to determine reliable methods, and will help with the application of laboratory quality assurance and control procedures.

Although a number of Member States have developed standard nuclear reference materials independently or in conjunction with other Member States, it is recommended that coordinated international efforts be undertaken to develop nuclear reference materials, both non-radioactive and radioactive. In Annex 3, a set of reference standards used in some countries is included. Inter-laboratory test programs (nuclear waste analytical round robins) should be developed and coordinated, focusing on analysis of raw waste, waste forms, and waste package reference materials. Round robins give the analytical laboratories the opportunity to evaluate their current analytical capability, test new analytical procedures, help improve analytical capability through a spirit of cooperation and learning at post round robin workshops, and provide traceable international analytical reference materials.

#### *3.2.5.2. Inter-laboratories collaboration and proficiency tests*

Accurate knowledge of the inventory of long lived and radiotoxic alpha, beta and gamma emitting radionuclides present in radioactive waste is a critical parameter for reliable classification [8] of the waste in compliance with the existing disposal site regulations. The activity limits for disposal of waste with long half-life and high radiotoxicity nuclides require having access to reliable characterization procedures, which provide results with a high accuracy.

The reliability of the final result at the end of the characterization process cannot be evaluated in a simple way. The uncertainty of this result is a function of its precision (repeatability, reproducibility) and accuracy (difference from the real value, expressed as the difference between a result (or mean) and the true value).

The precision of the result can be determined by an internal control in the laboratory using certified standards. However, the determination of the accuracy requires the following specific actions:

- Carrying out repeated analysis using different methodology, different analysts and different techniques,
- Carrying out control analysis with a reference matrix, or
- Participation in inter-laboratory comparisons (proficiency testing)

Inter-laboratory comparison studies can be a reliable way to determine the precision and accuracy of the results.

An intensive national and international collaboration among scientists, and the exchange of information on waste samples and radio-analytical methods among the different laboratories/institutions can help to identify the scope of application, the strengths and weaknesses of the methods and provide opportunities for improvement, harmonization, validation and finally qualification of selected methods within the scientific community. As an example, the European Commission promoted in the 1990s a Network [9] called EN-TRAP<sup>1</sup> devoted to join activities related the characterization and conformity of conditioned radioactive waste.

International proficiency tests will also lead to a higher confidence and acceptance of the applied procedures and of analytical results of each laboratory toward the institutions that have the responsibility of the disposal facilities, the Regulatory Body and other stakeholders. In this way, measurement and tests made in one country will be better accepted in another.

The International Union of Pure and Applied Chemistry has published a “Protocol for the design, conduct and interpretation of method-performance studies” [14]. Under the method proposed by this protocol, four steps are to be followed:

- Definition phase
- Design of the collaborative proficiency study
- Statistical analysis
- Final report

*a) Definition phase*

A collaborative study requires a special effort of definition, because this phase will establish the internal rules for comparison. Obviously these rules must be agreed upon by consensus and be fully accepted by all participants.

The objective of the study has to be clearly identified including estimates of the total within-laboratory uncertainty of the analytical results over the range of the values to be measured. The applicability of the methods to different types of materials or matrices, interferences, or whatever parameters affect the scope of the methodology have to be assessed.

The methods applied by each laboratory must be tested by internal and/or external protocols as well as sample preparation method, uncertainty assessment and calibration procedures.

The data and format of the results reported have to be defined in order to homogenize the data treatment in the inter-comparison test as well as the methods of accuracy determination.

Statistics play an important role in the comparison. The proposal presented below refers to DA of liquid radioactive wastes as proposed by IUPAC. Similar systems are available for NDA and are well explained in statistics books (see in Annex IV for references on statistics).

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<sup>1</sup> EN-TRAP: European Network of Testing Facilities for the Quality Checking of Radioactive Waste Packages  
[www.en-trap.org](http://www.en-trap.org).

*b) Design of the collaborative proficiency study*

The first step in the design is determining the number of materials to be tested in order to cover the range of application of the methods involved. The IUPAC's protocol indicates for a single type of substance, three to five test materials (samples) should be tested [14].

The number of replicates has to be established in the design phase to estimate the repeatability of the methods. The samples and replicates must be homogeneous in concentration levels of each analyte to be determined. The method to quantify the homogeneity has to be established in order to correct the differences for comparing the results. A good method to verify the homogeneity of the samples is to determinate the key or ETM nuclides.

Due to the special nature of the samples with radioactive material, difficulties or unaffordable economical costs may be encountered for transportation. In such a case, intercomparison tests should be substituted by intra-laboratory tests with an eye to repeatability of the measurements.

The concentration of certain nuclides in the samples may be a limitation for distributing homogeneous samples among the laboratories, due to radiation protection or regulatory limits of the facility in which the material will be analyzed. In this case, homogeneity checking is of particular importance.

A minimum number of aliquots and replicates have to be established in order to have sufficiently solid criterion for the statistical assessments of the proficiency test. Typically a minimum of three aliquots per sample are required for destructive analysis (with similar amounts of mass or volume); two of those are used (mineralized or prepared for measurement) for the required analyzes and the other one is kept as a reserve aliquot to cover any accidental event that results in a loss of information.

Each treated aliquot has to be divided into several - three is recommended - equal aliquots for performing the separation procedure for each nuclide, which means six (6) sub-aliquots per nuclide and laboratory.

An inactive waste sample or a simulant with the same physicochemical composition as the waste must be supplied to prepare blank samples to subtract matrix effects for each measurement and to perform a good estimation of the minimum detectable activity, to evaluate the analytical detection limits of each procedure.

To increase the reliability of the counting statistics, at least three replicates of each should be performed. In this way the counting value will be calculated through the arithmetic mean and the uncertainties through traditional statistical methods.

The process can be summarized in the following flow chart:

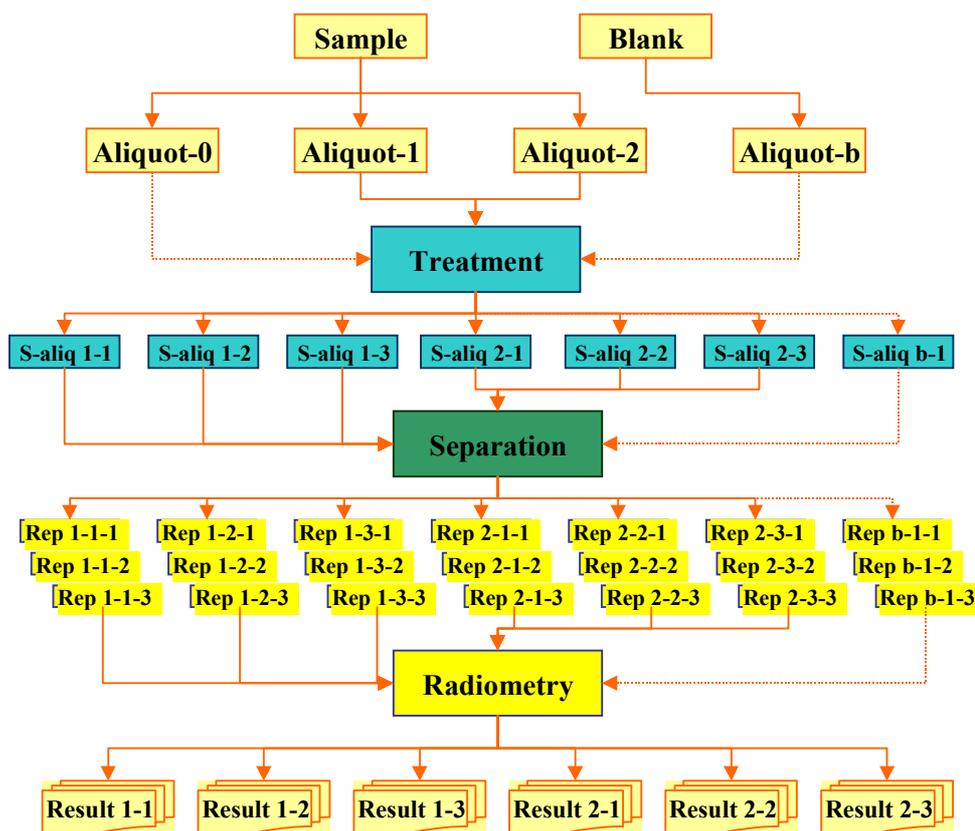


FIG 3.1 Scheme of sample aliquots and replicates

A data sheet has to be designed and distributed among the participants in order to organize the receipt of results and the communication between the evaluator and responsible scientists.

A code number for each laboratory, if necessary, will be assigned randomly to ensure the confidentiality of results in the evaluation and avoid the establishment of any kind of ranking among the participants.

The minimum contents of the data sheet have to be as follows:

— Identification data:

- ID -code of the LAB
- Responsible scientist
- Professional e-mail address
- Lab address

— Determination data per sample/nuclide

- Separation method (3 characters code)
- Radiometric method (3 characters code)
- Activity calculation equation
- Minimum detectable activity (MDA) equation

- Specific activity per sub-aliquot
- Specific MDA per sub-aliquot
- Type A uncertainty (see statistical meaning in Annex V)
- Type B uncertainty (see statistical meaning in Annex V)
- Uncertainty budget (see 3.4.)

c). *Statistical analysis*

Proficiency tests need a clear target established in the definition phase. If an artificial sample is prepared with standards, the results are easier to treat than in the case of real samples. However, in many cases artificial samples cannot be prepared with the same effect of matrix and interference as the real ones, so this method is recommended mainly for in-laboratory quality assurance.

In the case of non-synthetic samples, it is necessary to establish the way to evaluate the reference values (the better approach to the real value of radioactivity) and the target uncertainty (if the object of the inter-comparison is to get a maximum specific uncertainty in the determinations). They must be evaluated in two alternative ways:

- Consensus of expert or reliable laboratories. This option requires finding highly qualified labs that do not participate in the consortium and devoting a large budget for evaluating labs outside the consortium; and
- Consensus of participants to apply the statistical methods already existing. If this option is adopted, the evaluator has to take into account the restrictions and the limits that agreement values (the references values and/or target uncertainty agreed by consensus) means for statistical evaluation.

The evaluation of each individual Lab (i) data for each radionuclide (j) is done by the weighted average of six replicate values (m), which is the weighted factor of the reported combined (type A and B) variance ( $u^2(j_{X_m})$ )

$$j_{X_i} = \frac{\sum_{m=1}^6 \left( \frac{j_{X_m}}{u^2(j_{X_m})} \right)}{\sum_{m=1}^6 \left( \frac{1}{u^2(j_{X_m})} \right)}$$

if the uncertainty estimation is done in the right way (taking into account all relevant contributions) the uncertainty associated with this value is:

$$u(j_{X_i}) = \sqrt{\frac{1}{\sum_{m=1}^6 \frac{1}{u^2(j_{X_m})}}}$$

or if only type A uncertainty is evaluated the associated deviation is given by:

$$s({}^j x_i) = \sqrt{\frac{\sum_{i=1}^m \frac{({}^j x_m - {}^j x_i)^2}{u^2({}^j x_m)}}{(m-1) \sum_{m=1}^6 \frac{1}{u^2({}^j x_m)}}$$

The estimation of the population mean ( $\mu$ ) as the reference value of specific activity ( ${}^j X$ ) for each nuclide ( $j$ ) is calculated by the weighted average from data supplied ( ${}^j x_i$ ) by each Lab ( $i$ ) which is the weighting factor variance calculated for each Lab as above ( $u^2({}^j x_i)$ ).

$${}^j X = \frac{\sum_{i=1}^k \left( \frac{{}^j x_i}{u^2({}^j x_i)} \right)}{\sum_{i=1}^k \left( \frac{1}{u^2({}^j x_i)} \right)}$$

The uncertainty associated to this value is calculated by:

$$u({}^j X) = \sqrt{\frac{1}{\left( \sum_{i=1}^k \left( \frac{1}{u^2({}^j x_i)} \right) \right)}}$$

It is generally thought that results that significantly deviate from the corresponding reference values can be recognized as outliers by means of tests such as Cochran and Grubbs tests [14] [15]. Outliers flow is given in Figure 3.2.

Finally, the reference values are recalculated after the laboratories flagged by the mentioned procedure have been removed.

The first stage in the statistical evaluation of a proficiency test is to produce a score from a result  $x$  (a single measurement of analyte concentration in the material), obtaining an estimate of the bias ( $x-X$ ), with  $X$  the real value or the assigned value as the best estimate of the real value.

Proficiency test schemes proceed by comparing the bias estimate with a target value of standard deviation ( $\sigma$ ). The most important procedures for discussion in between laboratories for statistical evaluation of data are Q-score, z-score and u-score [16].

Q-score is based not on a standardized value but on the relative deviation

$$Q = \frac{x - X}{X}$$

This type of score relates to analytical error without any reference to a target quality value of  $\sigma$ .

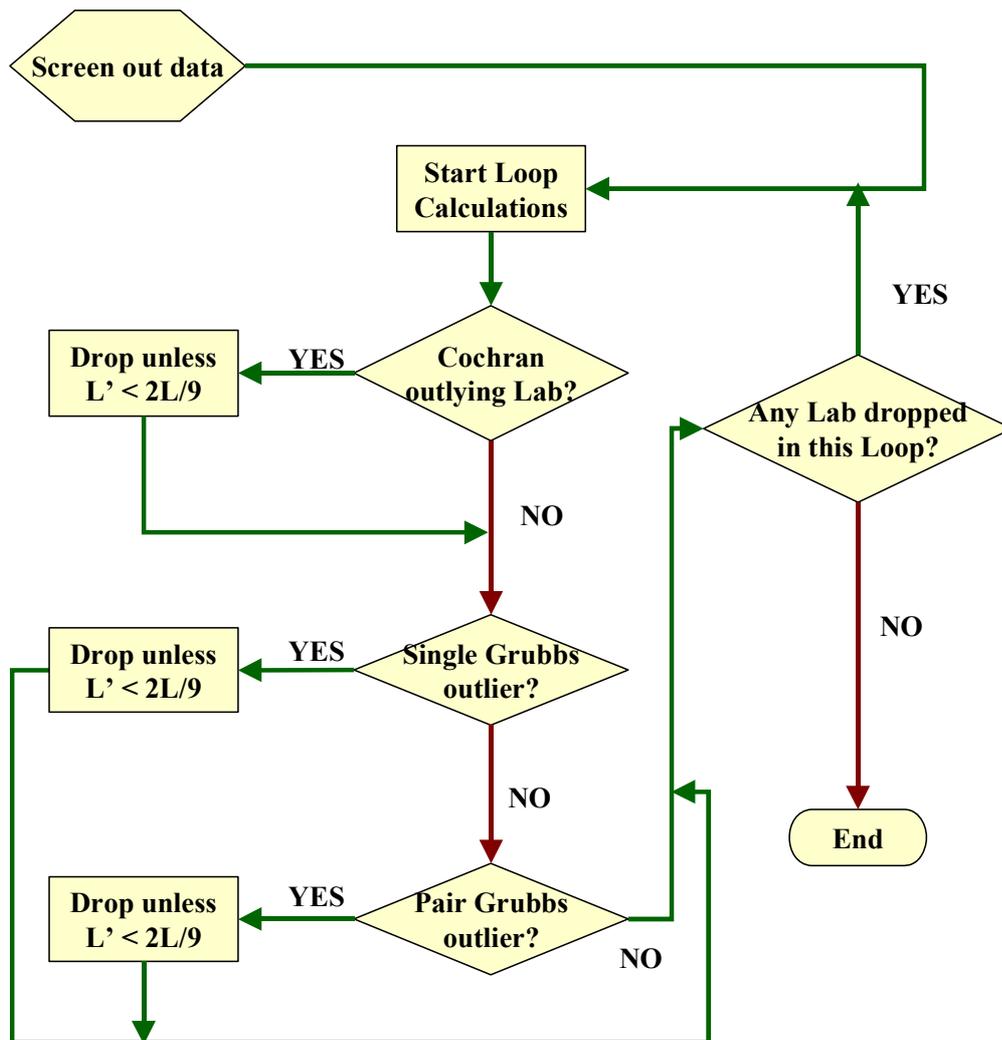


FIG. 3.2. Outliers flow.

Q-score measures the bias associated with the measurement, but the sensitivity to the outliers is rather low. It is necessary to examine the distribution of scores when defining criteria.

The criterion of performance called z-score captures an individual performance relative to the population's mean and variance.  $z = (X - M) / \sigma$  where:  $z$  refers to the z-score,  $M$  is the estimate of the population's mean,  $\sigma$  is the estimate of the population's standard deviation, and  $X$  is an individual score within the distribution having mean  $M$  and variance  $\sigma$ .

In the context of the inter-laboratory comparison, the equation is as follows:

$$z = \frac{x - X}{\sigma}$$

The target value for  $\sigma$  can be defined in several ways:

- By perception:  $\sigma$  is fixed arbitrarily
- By prescription:  $\sigma$  is an estimate of the precision required
- By reference to validate methodology: when a standard method is prescribed
- By reference to a generalized model: derived from a general model of precision

The z-score is standardized, so it is useful for a comparison between all analytes, test materials and analytical methods.

The meaning of the z-score can be immediately appreciated:

- $|z| \leq 2$ : Satisfactory
- $2 < |z| < 3$ : Questionable
- $|z| \geq 3$ : Unsatisfactory

Another type of evaluation is the use of the u-score, which is defined as the deviation of the reference value from the combined uncertainty between the uncertainty of the reference value calculation and the uncertainty of the individual determinations.

$${}^j u_i = \frac{{}^j x_i - {}^j X}{\sqrt{u^2({}^j X) + u^2({}^j x_i)}}$$

The advantage of this evaluation method relative to the others is that it does not require well-defined target values of the mean and standard deviation as in the case of the z-score. This statistical parameter follows a well-known distribution and is sensitive to the outliers in order to detect inconsistencies in the declaration of activity and uncertainties.

#### *d) Final report*

The final report must include a general description of the objectives of the proficiency test, confidentiality of the results and conditions and limits for the use of the data reported. The name of the participating organizations and responsibilities, the methods and materials tested and a detailed description of statistical evaluation methods (including symbols, definitions, etc), origin and calculation of reference data and their uncertainty as well as the outliers and their rejection criteria have to be included. Tables of individual data, the estimation of repeatability, results of the score that had to be met (q-score, z-score, u-score) in diagrams help clarify scientific evaluation of the results.

### **3.2.6. Consensus standards**

The reason for using voluntary consensus standards (materials as well as methods) is to produce comparable, accurate results regardless of laboratory variables. This can be accomplished through the use of standardized sample preparation, sample analysis and data reporting. The use of consensus standards is essential to ensure standardized, comparable results from all radioactive waste and simulant-based tests conducted within a project.

Typically, consensus standards are developed by private sector standards development organizations (SDO). These SDOs define and develop consensus standards using a process that encompasses several important characteristics. First, the standards definition and development process is open to all who are directly affected by the activity. Secondly, the standards committee, task or working groups must be comprised of representatives from all related groups of interest to the subject matter. These interests must be balanced, that is, producer, user, or regulatory groups cannot have the ability to ‘out vote’ any other group. And lastly, the standards development process must include a procedure by which individuals or organizations that believe the standard is technically incorrect, could cause potential harm, or

would impose undue hardship on any particular group have an opportunity for a impartial hearing of their concerns. In summary, consensus standards development organizations provide an open process whereby all interested parties and technical experts have the opportunity to participate in developing consensus in a fair manner.

To attain proper balance, this group must include experts on the subject from manufacturers, end users, research and independent testing laboratories, code authorities, regulators, government agencies and academics, that is, all those with a vested interest need to participate in the consensus process. Balanced membership within the group and adherence to a democratic process make it unlikely that views of one particular group would dominate. Instead, the process will likely foster development of technically sound standards that will address the needs of all concerned.

There are many benefits to using voluntary consensus standards. Harmonizing differing technologies and developing new technical standards allow for a common set of processes and results for comparison. Participation in the development process provides for effective communication between government and regulatory agencies and entities that must complete the work. The consensus standards development process provides a flexible environment in which the private sector can work together with regulatory and governmental agencies to meet the public goals of protecting the environment and health of the population. Participation in the process also helps to keep scientists and engineers technically current, promotes and preserves engineering and technological best practices, and avoids duplication of effort. Finally, bringing together experts from all groups with vested interests will result in consensus standards that meet the needs of private industry, government, and the public in a technically, environmentally, and economically sound fashion.

#### *Recommendation*

Participation in the consensus standards development and maintenance process, and use of consensus standards by all materially affected groups to embody an adequate set of standards for radioactive waste management, is essential. Government agencies and private industry must work together under a set of commonly developed consensus standards to meet the goals of regulatory policy and laws in a technically and economically sound manner. This will ensure that all goals pertaining to public health, safety, and protection of the environment will be met. Annex III provides a list of relevant standards along with an annotated bibliography that will aide those involved in radioactive waste characterization, conditioning, and management activities.

### 3.3. CALCULATION AND MODEL UNCERTAINTY

Whatever waste characterization strategy is adopted, the basis is to determine, by measurement or by modeling, the radiological, chemical and physical properties of the waste.

Most of these determinations are based on measurements, and the expression of a measurement is not complete with the value of only the measurand (the particular quantity subject to measurement), as it also requires the expression of its associated uncertainty.

The measurements on radioactive waste implies additional difficulties. For example, the complexity and heterogeneity of the matrices to be treated may lead to an inaccurate sampling

process and cause a large number of interferences in the analysis. In general, the uncertainty assessment process has to cover the following steps:

- Specification: write down a clear statement of what is being measured and the parameters on which it depends.
- Identify uncertainty sources: list sources of uncertainty for each part of the process or each parameter. Estimate the value of each uncertainty. At this stage, approximate qualitative values sufficiently; qualitative values can be refined in subsequent stages to provide the necessary level of quantitative values. Convert to standard uncertainty by expressing each component as a standard deviation. Calculate the combined uncertainty.

### *Specification*

The final aim of characterizing a certain waste stream is to determine the inventory of radionuclides from an ETM measurement and apply scaling factors between KN and DTM radionuclides. These scaling factors are based on the relationship between KN and DTM radionuclides ( $A_{DTM} = f(A_{key})$ , where the determination of  $A_{key}$  and the correlation between  $A_{key}$  and  $A_{DTM}$  have to be elaborated). For solid wastes, the determination of  $A_{key}$  can be performed by NDA on waste packages as described in Section 5.2 and Annex 1. The uncertainty associated with the radioactivity determination depends on the calibration methodology and knowledge of important parameters such as the distribution of radioactivity and density in the waste package.

In determining the scaling factors, additional elements have to be considered to define the uncertainty:

- Sampling of the primary waste. The homogeneity and representativeness of the radioactivity of the sample must be quantified.
- Radioactivity calculation of key nuclides. This process usually is performed by DA. It also avoids the use of transfer functions which are typical in NDA to correct important parameters, such as matrix composition, density and radioactivity distribution, etc. This way a more accurate input for scaling factor calculation will be provided.
- Radioactivity calculation of DTM nuclides. DA performs this process and the uncertainty associated is the one derived from each individual analytical procedure applied.
- Data correlation and fitting. This implies the contribution of the uncertainty of each input data ( $U_{KN}$  &  $U_{DTM}$ ) from sampling to the co-variance (*correlation* between two variables multiplied with the *standard deviation* of each) due to the data fitting.

Control of the sample homogeneity in the primary waste is not enough to assure a correlation between the activity measurement after packaging or conditioning and the activity calculated through scaling factors. A sample control at the conditioning level quantifies the homogeneity of the primary waste in the matrix (activity and density distribution).

### *Sources and types of uncertainty*

There are multiple sources of uncertainty and their identification, definition and quantification is one of the main tasks in the setting-up of a characterization process. The most relevant uncertainty sources in the activity determination are:

- Incomplete definition of the sample that will be analyzed

- Deviation or bias in the reading of an instrument
- Instrument resolution
- Value assigned to the reference data or parameters used in the calculations
- Representativeness of the sample measured
- Environmental conditions

According to the sampling process, the homogeneity of the samples must be considered in the uncertainty assessment of the performed determinations (representative ness). Besides the uncertainty associated with the sampling process in the analytical process, usually only the statistical uncertainty associated at the instrumental methodology (e.g. counting uncertainty and background or/and blank counting) is taken into account.

Two types of uncertainties were considered in the previous situation: random and systematic errors.

Random errors are obtained by statistical methods and consequently characterized by standard deviations, from a certain number of repeated measurements and from constant factors determined in the same way (e.g. calibration factor).

Systematic errors are due to physical effects influencing the measurement results. They cannot be evaluated by statistical methods. Systematic errors are expressed as limits of the mean value without an approach of the standard deviation value or another parameter that implies knowledge of the probability distribution. A former definition of systematic errors was the bias or the difference between the real value and the one obtained in the measurement. Their components are usually propagated linearly.

This classification is not clear and the final result of uncertainty depends on the analyst's consideration.

Nevertheless, the recommendations of the Bureau International de Poids et Mesures (BIPM) [28], as well as statistics books [15], establishes two types of uncertainty: A and B that only differ in the evaluation method and not in the origin of the uncertainty sources itself. In this sense, Type A uncertainties are the ones evaluated by statistical methods and Type B uncertainties are the ones evaluated by other methods [15].

Evaluation of type A uncertainty comes from maximum/mean contact dose rate and the calculation of apparent density. The evaluation of type B uncertainty arises from the deviation in the expected correction value and the real one in the assumption of matrix density distribution, activity distribution of each nuclide that contributes to dose rate and other relevant parameters such as the composition and thickness of inner shielding.

Since the type A uncertainty calculation is clearly described, a good evaluation of uncertainty type B leads to an appropriate accuracy of the results. Obviously the more complex the waste form is, the higher uncertainty the results will have.

### 3.4. UNCERTAINTY PROPAGATION

The expression of the measurement result needs a quality parameter with which the user of the data can estimate its reliability. The measurement of this quality parameter is the uncertainty of the result.

The uncertainty characterizes the distribution of values that can be reasonably attributed either to the measured item or to the value of the measurement. Results of the measurements would be useless if the uncertainty of the results cannot be combined or compared.

### *Uncertainty evaluation*

The method used for uncertainty evaluation has to be:

- Universal: applicable to every type of measurement and experimental data.
- Consistent: the value of uncertainty obtained has to be deductible from the components that contribute to the uncertainty and independent of the method associated.
- Transferable: the result has to be transferable; the uncertainty can be used as a component in the evaluation of the uncertainty of another measurement in which the result of the first measurement is used.

The BIPM recommendations [28, 29] define the combined uncertainty (see Annex V), as the one obtained by propagation of the variances assigned to the identified components of uncertainty and the expanded uncertainty as the one obtained by multiplying the combined uncertainty with a coverage factor ( $k$ ). Under the hypothesis of a normal distribution, confidence levels in function of  $k$  can be established in the usual way.

BIPM does not recommend the use of any particular cover factor and recommends that the analyst, characterization laboratory or organization defines the appropriated cover factor; nevertheless  $k$  must be specified in the expanded uncertainty.

The evaluation of Type A uncertainties is carried out through a statistical procedure valid for the experimental data management as follows (the degrees of freedom must be specified):

- Calculation of standard deviation and standard deviation of the average of a set of experimental values:
- Uncertainties obtained in the experimental data fitting to analytical functions by a least-squares method.
- Analysis of variance

The evaluation of type B uncertainties is carried out through all information available such as:

- Previous measurements.
- Experience on the properties of used instruments or materials.
- Specifications and manuals of the instrument manufacturer.
- Data from the instrument calibration.
- Uncertainties associated with reference data in handbooks or data evaluation sheets.

### *Sources of uncertainty in the characterization processes*

The aim of identifying the sources of uncertainty in the entire characterization process is to express mathematically the dependence of the final result on the input values and take into account all steps and assumptions made to reach the final result.

In case of destructive radionuclide analysis, several steps can be distinguished: sampling, sample preparation, separation process and radiometric process, and the calibration processes for the processes involving instrumental determinations.

The sampling process involves the estimation of the representativeness of the sample, usually depending on the homogeneity of the physical and radioactivity parameters (density, gross alpha, beta, gamma activity determination, etc) and expressing the representativeness as the standard deviation of the average of the controlled parameters.

Sample preparation and separation processes introduce uncertainties of the mass/volume determination used to calculate type B uncertainties. The uncertainty is usually calculated from the ratio of the concentration of a tracer or carrier by analytical measurements at the beginning and at the end of the process. This calculation is affected by measurement uncertainties and often by calibration uncertainties.

The radiometric process involves counting standard uncertainties, uncertainties of measurement efficiency, uncertainties of radioactive properties of the nuclide to be measured such as emission probability, decay constant, etc., and the uncertainties of calculation factors such as attenuation factors, self-absorption, counting geometry and dead time correction.

Calculating the uncertainty of the calibration process involves standard uncertainties of reference material counting, uncertainty of the certificate of the reference material, and the ones corresponding to other correction parameters of geometry, self-absorption, etc.

NDA process involves only sampling process, measurement process and calibration process. Sampling uncertainty can be calculated as in the case of destructive analysis by the representativeness of the sample, by the measurement of dose rate of the samples or by the calculation of total gamma activity by integral mode or open geometry.

The NDA measurement uncertainty consists of components from the determination of the net count rate at each energy, the efficiency of the equipment and other parameters such as the uncertainty corresponding to density calculation or transmission measurement for calculation of self-attenuation factors and/or attenuation factors, dead time correction factors, homogeneity of activity distribution, uniformity of density distribution and geometric correction factors.

#### *Combined & expanded uncertainty*

When the data  $x_i$  are not correlated, the combined uncertainty will be obtained according to the statistics expression:

$$u_c^2(y) = \sum_{i=1}^N \left( \frac{\partial f}{\partial x_i} \right)^2 u^2(x_i) = \sum_{i=1}^N (c_i u(x_i))^2 = \sum_{i=1}^N u^2(y_i)$$

The terms  $\left( \frac{\partial f}{\partial x_i} \right)$  are called sensitivity coefficients ( $c_i$ ) and describe the variation of the estimated value ( $y$ ) as the components ( $x_i$ ) vary.

If the data are correlated, the combined uncertainty is calculated as:

$$u_c^2(y) = \sum_{i=1}^N \sum_{j=1}^N \left( \frac{\partial f}{\partial x_i} \right) \left( \frac{\partial f}{\partial x_j} \right) u(x_i, x_j) = \sum_{i=1}^N \left( \frac{\partial f}{\partial x_i} \right)^2 u^2(x_i) + 2 \cdot \sum_{i=1}^N \sum_{j=1}^N \left( \frac{\partial f}{\partial x_i} \right) \left( \frac{\partial f}{\partial x_j} \right) u(x_i, x_j)$$

The final step is to multiply the combined standard uncertainty by the chosen coverage factor in order to obtain an expanded uncertainty. The expanded uncertainty is required to provide an interval, which may be expected to encompass a large fraction of the distribution of values that could reasonably be attributed to the sample [16]. The expanded uncertainty according to the BIPM recommendations is expressed as follows:

$$U = k \cdot u_c(y)$$

The selection of the cover factor (k) depends on the desired confidence level associated with the interval. It is desired to assign a well-defined confidence level to each value of k, but if the probability distribution is not well known this is not possible.

### *Uncertainty budget*

The uncertainty budget is an instrument to compile and record the step-by-step procedure for calculating the uncertainty. It can be summarized as:

- To express mathematically the dependence of the sample on the input data
- To establish the relation amongst all uncertainty sources
- To calculate the type A uncertainties
- To calculate the type B uncertainties
- To calculate the contribution of each type of uncertainty to the total combined uncertainty, applying the corresponding expression, if necessary with co-variance.
- Select a cover factor k and calculate the expanded uncertainty.

The management of all uncertainties involved in a characterization process values and parameters can be a source of mistakes. To prevent this, it may be useful to set up an uncertainty budget per individual process.

Two types of uncertainty budget can be defined: the absolute uncertainty budget, involving all uncertainties and their characteristics that will be related by mathematical summation ( $y = x_1 \pm x_2$ ) if the uncertainty is easier to calculate by propagation of individual absolute uncertainties, and the relative uncertainty budget, that depends on the multiplication of input values ( $y = x_1 * / x_2$ ) if it is easier to calculate the relative combined uncertainty from individual relative uncertainties of each input values. Tables II and III show possible absolute and relative uncertainty budgets, respectively.

The input fields recommended for the absolute uncertainty budget are the symbol of the input value  $x_i$ , the input absolute uncertainty  $\Delta x_i$  in the same units as  $x_i$ , a description of the input absolute value given (e.g. limits or standard uncertainty, etc), probability distribution assigned to the variable, the divisor ( $d_i$ ) (that depends on the probability distribution associated to the variable  $x_i$ ) and the information available regarding to the uncertainty of the value  $x_i$  (e.g. if  $\Delta x_i$  of a certain value is  $3\sigma$  of the value  $x_i$ , the divisor of  $\Delta x_i$  will be 3) to

convert  $\Delta x_i$  in  $u(x_i)$  the value of  $u(x_i)$  as  $\Delta x_i/d_i$ , The absolute sensitivity coefficient  $c_i$  and finally the value of  $u_i(y)$  calculated as  $c_i \cdot u(x_i)$  expressed in  $x_i$  units.

TABLE II. ABSOLUTE UNCERTAINTY BUDGET

Symbol	Value $\pm$	Where the value given describes	Probability distribution	Divisor ( $d_i$ )	$U(x_i)$	$c_i^{abs}$	$u_i(y)$ $\pm$

The input fields recommended for the relative uncertainty budget are the symbol of the input value  $x_i$ , the input relative uncertainty given by  $(\Delta x_i \cdot 100)/x_i$  in %, a description of the input uncertainty value given, probability distribution assigned to the variable, regarding to the probability distribution and information available in order to convert  $\% \Delta x_i$  in  $\% u(x_i)$ , the value of  $(u(x_i) \cdot 100)/|x_i|$  in % as  $(\Delta x_i \cdot 100)/(x_i \cdot d_i)$ , the relative sensitivity coefficient  $c_i$  and finally the value of  $\% u_i(y)$  calculated as  $c_i \cdot (u(x_i) \cdot 100)/|x_i|$  that it will be expressed in % of  $x_i$ .

TABLE III. RELATIVE UNCERTAINTY BUDGET

Symbol	Value $\pm \%$	Where the % value given describes	Probability distribution	Divisor (Div)	$U(x_i)$ $\pm \%$	$c_i^{rel}$	$u_i(y)$ $\pm \%$

An example of a complete uncertainty calculation process of a characterization procedure is given in Annex V.

## 4. CHARACTERIZATION METHODOLOGY APPLICATION TO THE DIFFERENT WASTE CATEGORIES

### 4.1. INTRODUCTION/APPLICABILITY OF SCALING FACTOR METHOD

It is broadly accepted that the vast majority of waste characterization applications will involve the use of scaling factors to some degree. For all but the most simple cases, there will be some DTM or ITM nuclides that must be assessed in a practical way. For example, disposal of spent sources with traceable calibration certificates will not require the use of scaling factors, but most other cases will. It must be recognized that the use of scaling factors is a serious and complex matter. There are many pitfalls and it is easy to be misled. The following general guidance addresses many of these potential pitfalls.

1. Changes to chemistry or process is likely to change the distributions of nuclides, so scaling factors will need to be reassessed following any significant change, for example, changes to reactor power rating will change burn-up profile, changes to process chemistry, resin or filter types, etc.

2. Scaling factors require a technical basis for the existence of a correlation. If there is not a technical basis for the correlation, it may not be valid or defensible in practice. Especially beware the “log-log trap”. If logarithmic regression is necessary to produce a mathematical correlation, the uncertainty grows dramatically and may become several orders of magnitude. Although a correlation may visually appear to be present on a log-log plot, it is necessary to consider the spread of data points or variance. Although it may not appear to be visually large, graduations on a log plot are orders of magnitude. It may be difficult to justify that a true correlation exists if the variance is several orders of magnitude.
3. Scaling factors are waste stream and case specific, not reactor type specific. As relative distributions of nuclides in wastes are highly process and chemistry dependent, it is uncommon for generic scaling factors to be developed, although it is recognized that the existence of such generic sets would be highly desirable to a waste manager. Where generic scaling factors are used, they usually are applied to only a few specific pairs of nuclides. It is necessary that all scaling factors are demonstrably valid and the uncertainty for the specific case has been assessed (see Section 3). Before using any generic set of scaling factors, it would be necessary to demonstrate that the plant conditions fall within the bounds of conditions that led to the development of the set. For the set to be truly representative, the population assessed would have to exhibit similar design materials, process history and process chemistry. If there are many variables, then there may not be an underlying technical basis for the correlation (see point 2 above). In addition, with large populations, the resultant uncertainty may be very large. This is not to say that a generic set of scaling factors could never be valid, but it would have to be assessed and shown to be valid prior to use.
4. It is common practice when developing scaling factors or fingerprints to pre-survey the stream to identify regions of elevated activity. Samples are taken specifically from this upstream area to ensure good quality statistics of the ratios of activities of the various nuclides (i.e., you need radioactivity to determine a ratio, and the more radioactivity you have, the more accurate the ratio will be). This initial scoping is often most practically performed using gamma spectroscopy. There are a few rare occasions where this approach can miss entirely some DTM radionuclides. This situation occurs when DTM nuclides are separate, or become separated and exist in large concentrations in the absence of associated ETM radionuclides. Process chemistry can lead to this situation, but it is more common in laboratory situations, where laboratory processes are designed to separate sample constituents. Where it is possible for this to occur, a combination of pre-survey techniques should be employed to identify areas of elevated activity – for example, use of both gamma and beta surveys will identify areas of significant pure beta radionuclides activity. An example of this situation is decommissioning a laboratory facility. Another specific nuclide where this can be a particular problem is tritium. Due to its extremely high mobility, it is often encountered on its own (i.e., without a corresponding scalable nuclide present). Due to its very low energy beta emission, it is very difficult to detect. Where tritium is likely to be present, great care must be taken in developing an appropriate scaling factor.
5. The national (or commercial) policy regarding whether to use mean values or conservative values for scaling factors will have to be identified. While there is a sound scientific basis for using mean values as the overall inventory of the repository will be more accurate, it may not be permitted in some jurisdictions to allow underestimating of

any nuclide concentrations. On the other hand, if the most conservative values for the scaling factors based on the log-log plots were used, a situation might be achieved where the real radioactivity in a repository is much smaller than the declared one – resulting in much higher and superfluous costs. In the case where wastes are to cross international borders, it will be necessary to know the position in all countries involved. This is also a consideration when uncertainties may be very large.

6. There is a trend in certain organizations to use scaling factors to assess ETM nuclides in addition to DTM and ITM. In principle, this is not necessary because ETM nuclides can be directly measured. However, there may be particular advantages for this approach. It is necessary to fully understand the uncertainty implications of both approaches and it is recommended to use the approach with the smaller uncertainty. In addition, it is advantageous to periodically use the alternative approach and compare results to continuously confirm the validity of the scaling factors.
7. If scaling factors are used to assess DTM and ITM in waste or waste forms, it is important to validate these factors through DA. This is specially so for the radionuclides affecting the long term dose to man during the surface or geological disposal of the waste packages – e.g. I-129 and Se-79 in case of geological disposal of high-level waste.
8. When using analytical data on small samples to calculate scaling factors, it is highly important to upscale to full size drums. This is especially true in case of variable waste streams. A sufficiently large set of samples should be taken and analyzed according to a sampling strategy. Non-destructive analysis on the full size drums will be helpful to verify the scaling factors.

## 4.2. SIMPLE AND STABLE WASTE STREAMS

### 4.2.1. Enrichment, conversion, fuel fabrication

Fuel cycle operations can be performed using a variety of fabrication processes. Enrichment can be carried out by centrifugation or by gaseous diffusion, and fuel fabrication may involve different methods or different fissile materials (e.g. MOX fuel).

It would be unrealistic to specify recommendations applicable in all circumstances. Two examples are cited to illustrate what could be an acceptable methodological approach for declaring the radionuclides found in the waste packages. Each participant in the process (waste producer, regulatory authorities, and organization responsible for ultimate disposal) is responsible for adapting these examples to other comparable situations.

#### 4.2.1.1. *Natural uranium enrichment by gaseous diffusion*

The gaseous diffusion process for enrichment of natural uranium uses uranium hexafluoride  $UF_6$ . Uranium is a mixture of three natural isotopes, whose molar concentrations—and therefore whose relative radioactivities—vary throughout the diffusion cascade.

In contact with water vapour, even in trace amounts,  $UF_6$  hydrolyses to form aerosols that are deposited on surfaces encountered by the fluid, resulting in radioactive surface contamination. All tubes and vessels in contact with the process fluid are contaminated to some extent.

Maintenance and servicing operations in the enrichment plant or facilities used for disassembly, cleaning, adjustment, repair, replacement, etc., thus produce contaminated waste and residues that must be collected and eliminated.

A distinction is made between compactable and non compactable waste, and between wet and dry waste, by source segregation.

Production is continuous, although a few interruptions occur for some types of plant components (e.g. valves, pipes, rotating machinery, etc.) during preventive maintenance activities.

#### *General waste characteristics*

Compactable waste mainly consists of materials used for the following:

- in situ decontamination of parts after disassembly or maintenance of process tubes and equipment; these materials are mainly rags, cotton swabs, synthetic absorbent fabrics;
- protection of surfaces against contamination during maintenance operations: PVC or polyethylene sheeting and bags;
- protection of personnel against contamination: gloves, clothing, PVC or polyethylene;
- treatment (soaking) of filter canisters and permanent or mobile purification devices; the latter include a combination of filter, rubber gasket, PVC, etc.

A mean physical composition can be calculated for the compactable waste (percentage of PVC, polyethylene, latex, paper, cotton, fabrics, etc).

Non compactable waste consists mainly of scrapped parts and materials:

- metal, rubber or synthetic gaskets,
- complete or cut-up metal parts that would be significantly more expensive to decontaminate than to ship to a disposal site,
- absorbent materials,
- rubble,
- soil.

The traceability of the non compactable waste form placed in each drum must be ensured.

#### *Nature and activity of the radionuclides*

Contamination in the waste consists primarily of aerosols or fine particles deposited on the previously mentioned physical surfaces after contact with the atmosphere or other surfaces. It can also be due to impregnation after contact with a liquid. The contamination is thus generally not strongly bound to the surface. The contaminating agent is uranium:

- mainly as uranium oxyfluoride ( $\text{UO}_2\text{F}_2$ ) resulting from hydrolysis of  $\text{UF}_6$ ; the oxyfluoride may be anhydrous but is generally found as  $(\text{UO}_2\text{F}_2, x\text{H}_2\text{O})$  or even  $(\text{UO}_2\text{F}_2, x\text{H}_2\text{O}, y\text{HF})$ ;

- the form of complex fluorides produced by the reaction of  $UF_6$  with fluids such as oil or with gaseous chlorofluoride compounds.

While oxyfluorides are highly soluble in water, acidic and basic solutions, the products of the second category are very often insoluble.

#### *Methodology for declaring the radionuclides in waste materials*

Uranium is a mixture of three natural isotopes (234, 235 and 238); the  $^{235}U$  concentration varies across the diffusion cascade.

A mean uranium-235 concentration can be determined based on the experience acquired over several years of plant operation. The declaration must be thoroughly prepared on the basis of representative waste samples from all the process facilities taken at frequent intervals throughout the year. In this context, the results of sample measurements of gaseous discharges from the plant to the atmosphere would appear to be suitable indicators.

Having evaluated the mean  $^{235}U$  concentration, the corresponding  $^{234}U$  concentration must then be calculated; the remainder represents the  $^{238}U$  concentration.

The mean molar isotopic composition of the representative waste mixture is used to calculate the specific activity of the mixture and to declare the radioelements accompanying uranium. Uranium is always found together with its short half-life decay products,  $^{234}Th$  and  $^{234}Pa$ , the first daughter isotopes of  $^{238}U$ .

These radioisotopes can be present in the waste:

- either in nominal proportions with respect to uranium—i.e., at radioactive equilibrium—when the contamination is due exclusively to uranium,
- or in slight excess, when the contamination arises mainly from nonvolatile Th and Pa volatile fluorides that generally deposit at the bottom of process vessels or at singularities in the flow lines.

In the second case, once they are isolated, the activity of the fluorides decreases at the half-life of  $^{234}Th$  (24 days); i.e., between the moment the waste is produced and the moment it reaches the disposal site, the  $\beta\gamma$  activity is no longer significant.

The radioactivity of the waste package is calculated:

- from  $\gamma$  spectrometry measurements (on the principal  $^{235}U$  line) for each package shipped,
- from a mathematical transfer function allowing for the influence of the parameters likely to affect the measurement (weight, type and density of the waste, measurement distance),
- from the calculated standard spectrum ( $^{234}Th$ ,  $^{234}Pa$ ,  $^{238}U$ ).

#### 4.2.1.2. MOX fuel fabrication

##### *General methodology*

In a plant fabricating MOX fuel assemblies for nuclear power stations, production is organized in fabrication campaigns to supply the client with fuel assemblies meeting the nuclear specification requirements (Pu and U isotopic composition and content at a given reference date).

The finished product is a mixture of uranium and plutonium with well-defined characteristics, making it a simple matter to calculate the standard spectrum for each production campaign. The reference spectrum is thus the standard spectrum for the production campaign in progress at the moment the waste containers are filled. This data must be conserved for traceability purposes.

If the waste arises from a zone in which only one of the two constituents is handled (e.g. the uranium zone), it is advisable to specify the standard spectrum for that zone and declare the radionuclides in the waste accordingly.

The standard reference spectrum is validated on the following assumption. If the process used to produce fuel rods with the contractual nuclear characteristics is fully controlled, there is a strong probability that the waste produced will have identical characteristics. Consequently, in routine production the waste drums are measured, and the radionuclides declared are those identified in the standard reference spectrum. Conversely, additional measurements are required in the event of a malfunction liable to affect the standard reference spectrum.

In this type of production the beta-emitting radionuclides spectrum is often simple (e.g.  $^{99}\text{Tc}$ ) and can be determined with respect to the total alpha activity rather than with respect to a single key nuclide.

##### *Standard spectrum management*

When the alpha activity (at the reference date) of the MOX fuel varies minimally from one production campaign to the next (this criterion must be the subject of a common agreement between the waste producer, the regulatory authority and/or the organization responsible for final disposal), the standard reference spectrum of the preceding campaign can be reused.

This procedure allows successive production campaigns to be organized at optimum cost without significantly affecting the quality of the data characterizing the waste.

#### **4.2.2. Nuclear power plant**

This waste results from treatment of cooling water, equipment decontamination, and routine facility maintenance. As discussed in Section 3, part of the traceable waste streams from NPPs can be classified as *simple and stable*. The main characteristics of this type of waste are:

- sampling is easy or not necessary
- easy to measure using NDA or DA methods
- simple spectrum
- good relation between dose rate and activity (scaling factor method is applicable)
- physical and chemical properties are known or easy to measure (DA)

Typical examples of this category are filters, neutron sources, calibration sources, and defectoscopy sources.

An ISO standard guide “Scaling factor to determine the radioactivity of low and intermediate radioactive waste packages generated at NPP” [18] is in progress. The contents of this guide describe the principles of the scaling factor methodology applied to NPP, the sampling requirements (representation, rejection of outliers, records of samples) and the evaluation methodology for scaling factors (evaluation by linear or non-linear relationship; selection of key-nuclides; integration of corrosion products, fission products and alpha emitting nuclides, and accuracy). This scaling factor methodology for NPP operational waste is suitable when many samples can be taken from the same waste stream. This methodology is not applicable to current reprocessing waste, because this typically involves many different campaigns.

#### **4.2.3. Spent fuel**

Spent fuel actually consists of two barriers that may prevent release of radionuclides: the cladding and the fuel matrix itself. Some programs take credit for the protection cladding may offer. However, other programs do not consider cladding as a barrier mostly because the final state of the cladding after irradiation is not always well known. For example, cladding failures in the fuel can usually be identified by radioactivity release to the reactor coolant during cool down and depressurization of the reactor. While this will identify the presence of failed fuel, additional techniques such as sipping are sometimes needed to accurately identify the failed assembly. Still, it might not be known how many rods within that assembly have failed. The presence of tramp material in the core may also make it difficult to detect pin-hole defects in cladding. A combination of sipping, visual inspection, eddy current or ultrasonic testing may be performed to identify the percentage of failed fuel rods.

Even if the number of failed rods can be accurately determined and documented, it is difficult to know the state of intact fuel rods. During reactor operation, the cladding undergoes oxidation and hydriding that can affect their performance. As the fuel is pushed to higher burnups, these reactions become more significant. Thus, it is not clear what the remaining wall thickness is for the intact rods. The uncertainty in how many additional rods might fail during handling, transportation, or storage due to these incipient failures is the main reason many programs do not take credit for cladding as a barrier to radionuclide release.

Once the cladding is breached, fission gases and other volatiles in the fuel/clad gap and plenum may be released. The fraction of inventory available for release is a function of fuel burn-up and the power (temperature) history. This can be readily calculated using records from the utility and codes such as ORIGEN. Additional radionuclides can then be released if the fuel is exposed to oxidizing conditions either by air or water or even as result of radiolysis under otherwise reducing or anoxic conditions.

## 4.3. COMPLEX AND STABLE WASTE STREAMS

### 4.3.1. Nuclear power plant

As discussed in Section 3, part of the traceable waste streams from NPPs can be classified as *complex and stable*. Main characteristics of this type of waste are as follows:

- sampling is easy but must be representative (for instance, the total radioactivity in ion exchange resins may not be stable with time)
- fingerprint is known and stable
- matrix and origin of waste is known
- easy to measure using NDA or DA methods
- scaling factor method for difficult to measure radionuclides is applicable
- physical and chemical properties are easily determined by destructive analysis

Examples of this category include evaporated concentrates, ion exchange resins and solid waste.

### 4.3.2. Reprocessing

#### 4.3.2.1. Subject and scope

The scaling factor methodology is applicable to nuclear facilities where complex radioactive substances) are handled, for instance, a reprocessing plant. It is further applicable if a fraction of the radionuclides present in the mixture can be assayed directly by non-destructive measurement such as gamma spectrometry, dose rate measurement and passive or active neutron counting. A list of radionuclides likely to be found in the initial reprocessing sludge is given in Annex II.

The activity of DTM and ITM radionuclides (called DTM in this Section) is determined from the activity of the directly measurable radionuclides by means of prior correlations established either from destructive analysis or using calculation codes.

#### 4.3.2.2. General application criteria of the scaling factor methodology

From the standpoint of radionuclide behaviour, the physicochemical processes implemented in a reprocessing nuclear facility can be represented schematically as follows:

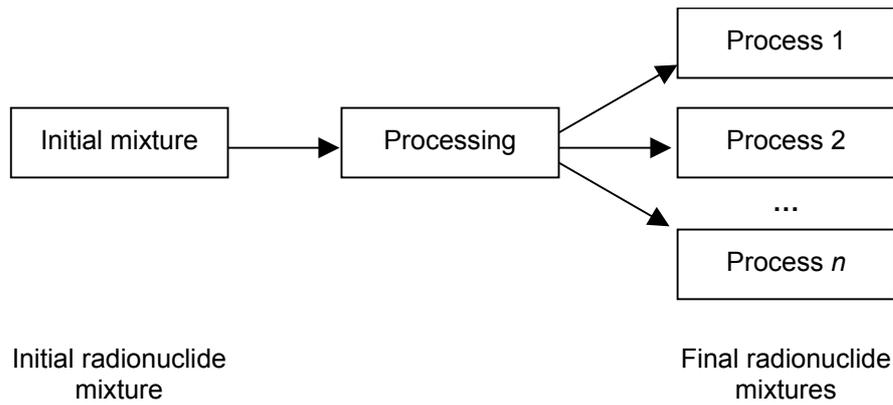


FIG 4.1. Physicochemical processes in a reprocessing facility.

The physicochemical process may:

- modify the isotopic ratios of a single chemical element (e.g. enrichment);
- conserve the isotopic ratios of the chemical elements but modify the chemical element concentrations in the initial solution.

The following values are defined:

- the decontamination factor (DF) of a radionuclide is equal to the ratio of the activity of the radionuclide in the initial mixture to its activity in the final mixture:

$$DF = \frac{\text{Initial activity}}{\text{Final activity}}$$

The DF is a mass or volume factor, depending on whether the initial and final activity is expressed in terms of the mass or volume of the initial and final mixtures;

- the reconcentration factor (RF) is equal to the ratio of the activity of the radionuclide in the final mixture to its activity in the initial mixture:

$$RF = \frac{\text{Final activity}}{\text{Initial activity}}$$

It is equal to the inverse of the decontamination factor:  $RF = \frac{1}{DF}$ .

#### 4.3.2.3. Selection criteria for Key Nuclides (KN)

To establish representative correlations, KNs are selected according to the following criteria:

- the KN must be present in the assay sample in significant quantities;
- the radioactive half-life of the KN must be long compared with the time necessary for sampling, measurement, and physicochemical processing;
- the KN and DTM in the initial mixture should preferably be formed by similar mechanisms. For example, in nuclear power plants, the radionuclides arising from fission reactions in the fuel should be distinguished from radionuclides due to activation of structural materials;
- the chemical behaviour of the KN and DTM in the reprocessing steps should be similar; it is thus preferable to choose isotopes of the same chemical element.

#### 4.3.2.4. *Establishing correlations between KN and DTM in the initial mixture*

##### **Technique based on sampling and destructive measurement**

A representative sample is taken in the initial mixture. The laboratory analysis procedure implements techniques capable of quantifying the activity of the desired radionuclide(s) in the prepared sample.

In a complex sample the analysis may reveal a non linear relation between the activities of the KN and DTM. In most cases this relation can be reduced to the following expression:

$$\text{Activity}_{\text{DTM}} = a \cdot (\text{Activity}_{\text{KN}})^b$$

The above activities refer to the measured specific activities or volume activities. The terms a and b are defined by fitting the preceding relations to the experimental values using suitable mathematical methods (e.g. least-squares method) to minimize the deviation.

##### **Technique based on calculation codes**

The radionuclide activities in the initial mixture can in some cases be determined using specific calculation codes such as the following:

- fuel evolution codes for nuclear facilities in the back end of the fuel cycle;
- activation codes in facilities concerned by material irradiation.
- 

The correlation is expressed by a ratio:  $\frac{\text{Activity}_{\text{DTM}}}{\text{Activity}_{\text{KN}}}$ .

The above activities refer to the measured specific activities or volume activities.

When the activity of a DTM can be correlated with the activity of several KN, the characteristic correlation ratio can be established from the sum of the KN activities:

$$\text{Correlation ratio} = \frac{\text{Activity}_{\text{DTM}}}{\sum \text{Activity}_{\text{KN}}}$$

#### 4.3.2.5. Establishing correlations in the final mixture

##### Technique based on sampling and destructive measurement

The technique discussed in 4.3.2.4. is also applicable when quantifying the radionuclides in the final mixture with the same constraints as for the initial mixture.

##### Technique based on knowledge of the characteristics of the initial mixture and of the physicochemical treatment applied

The correlations between KN and DTM in the initial mixture are known. The effects of physicochemical processing on the radionuclides concentrations have first to be characterized by destructive analysis (the DF or RF values of the radionuclides are known).

By defining the decontamination or reconcentration factors, the following relations can be established:

$$\begin{aligned} \left( \frac{\text{Activity}_{\text{DTM}}}{\text{Activity}_{\text{KN}}} \right)_{\text{final mixture}} &= \left( \frac{\text{Activity}_{\text{DTM}}}{\text{Activity}_{\text{KN}}} \right)_{\text{initial mixture}} \times \left( \frac{\text{DF}_{\text{KN}}}{\text{DF}_{\text{DTM}}} \right) \\ \left( \frac{\text{Activity}_{\text{DTM}}}{\text{Activity}_{\text{KN}}} \right)_{\text{final mixture}} &= \left( \frac{\text{Activity}_{\text{DTM}}}{\text{Activity}_{\text{KN}}} \right)_{\text{initial mixture}} \times \left( \frac{\text{RF}_{\text{DTM}}}{\text{RF}_{\text{KN}}} \right) \end{aligned}$$

If a DTM is correlated with several KN in the initial mixture, only the directly measurable radionuclides with non-zero reconcentration factors (and with a specified decontamination factor) will be used as tracers for the final mixture.

If the following relations occur:

- $A_{\text{DTM init}}$  and  $A_{\text{DTM final}}$  are the specific and volume activities of the DTM in the initial and final mixtures;
- $A_{\text{KN init}(i)}$  and  $A_{\text{KN final}(i)}$  are the specific and volume activities (subscripted i) of the n KN in the initial and final mixtures with non zero reconcentration factors;
- $\text{DF}_{\text{DTM}}$  and  $\text{RF}_{\text{DTM}}$  are the mass or volume decontamination and reconcentration factors for the DTM; and
- $\text{DF}_{\text{KN}(i)}$  and  $\text{RF}_{\text{KN}(i)}$  are the mass or volume decontamination and reconcentration factors (subscripted i) of the n KN.

then the correlation between the sum of the activities of the key radionuclides and the activity of the DTM in the final mixture will be as follows:

$$\frac{A_{\text{DTM final}}}{\sum_{i=1}^n A_{\text{KN final}}} = \frac{A_{\text{DTM init}}}{A_{\text{KN init}(i)}} \cdot \frac{\text{DF}_{\text{KN}(i)}}{\text{DF}_{\text{DTM}}} \cdot \frac{1}{1 + \sum_{i \neq j=1}^n \frac{A_{\text{KN init}(i)}}{A_{\text{KN init}(j)}} \cdot \frac{\text{DF}_{\text{KN}(j)}}{\text{DF}_{\text{KN}(i)}}}$$

The expression with the reconcentration factor RF is determined from the above expression by substituting  $1/RF$  for DF. The KN subscripted (j) in the preceding expression is generally the KN with the most significant final specific activity or volume activity and the most easily measurable by the method used.

### 4.3.3. Waste form

#### 4.3.3.1. Introduction

Conditioning is defined as those operations that produce a waste package suitable for handling, transport, storage or disposal. Conditioning may include converting the waste to a solid waste form, enclosure of the waste in containers, and, if necessary, providing an overpack [17]. In some cases, the waste may be immobilized into a typical fixation matrix (for example, bitumen, cement or glass). This category of waste is well characterized, with a stable and known fingerprint, knowledge of chemical and physical parameters, radiation stability, and long term behaviour. Classification of this waste is *complex and stable*.

Main characteristics:

- stabilized waste form (physical and chemical properties are stable and known)
- stable and known fingerprint
- scaling factor method for activity determination is applicable
- radiochemical sampling is recommended but not strictly required (process and final product control should be sufficient along with possible NDA)
- physical and chemical property measurement will use DA (leaching, compressive strength, penetration test, etc.).

Examples: cemented, bituminized, vitrified products.

#### 4.3.3.2. Vitrified waste

For most Member States, the reference waste form for high level waste generated from reprocessing of spent fuels is borosilicate glass [19]. Reprocessing wastes are often compositionally complex due to the variety of spent fuels, complicated chemical processing schemes, and storage schemes used. Due to waste composition complexity, glass composition formulation must be optimized to meet the often competing property and composition constraints, such as waste loading and cost, processing rate, viscosity and electrical conductivity of the melts, melter corrosion, phase stability, chemical durability, and regulatory compliance. However, a well-designed waste conditioning process produces homogeneous glass within a composition range in which the property distributions do not vary significantly, allowing for relatively easy representative sampling if required.

With enough initial non-radioactive and radioactive characterization work, key waste glass properties can be predicted as functions of composition (e.g. for a qualified glass composition region within a narrow composition range, most glass properties can be accurately modelled as a linear function of composition, i.e., stable). In this situation, waste characterization data generated during the conditioning process can be used for process control and compliance activities [see references in Annex IV (Section 4) for glass composition versus property modeling]. If the spent fuel parameters, such as initial fuel mean

composition, neutron flux and burnup and conditioning process, are sufficiently known, it has been demonstrated that fission yield scaling factors can be determined to predict the concentration of fission products and actinide radionuclides for repository acceptance of the vitrified waste [20]. This determination of the scaling factors is based on DA results on HLW samples.

Additionally, the use of fission yield scaling factors may be beneficial in controlling or understanding the conditioning process via mass balance closure. For example, one could measure the radionuclides contained in the glass and recognize that certain volatile components were not in the correct ratio and know that they need to be accounted for within the off gas system [21]. This facilitates a scheme of minimal to no waste glass sampling and measurement, given that destructive chemical analysis of glass is costly and hazardous due to the high concentration of radionuclides.

#### *4.3.3.3. Cemented and bituminized waste*

Cement solidification for complex and stable waste streams is typically used for NPP operational waste, such as ion exchange resins, evaporator concentrates, and precoat filter sludges. Cement is also used to solidify intermediate level reprocessing sludges. The cement matrix may be the most common ordinary Portland cement (OPC), more durable cements (blast-furnace slag cement, others), or include admixtures. Important considerations in the cementation process are (1) that the cement formulation and the waste must be compatible, (2) the cement-waste mixture must be sufficiently homogenized during the cementation, and (3) that the compressive strength of the cemented waste is sufficient, requiring a minimum curing period [22, 23].

Bituminization is applicable to roughly the same types of NPP operational waste as those that are currently being cemented. Types of waste suitable for immobilization in bitumen are sludges and slurries, ion exchange materials, liquid concentrates, and incineration ashes. Intermediate level reprocessing precipitates are also known to be bituminized. The two types of bitumen mainly used are distilled or blown bitumen, depending on their production process. Blown bitumen is harder than distilled bitumen. Important considerations in the bituminization process are (1) that the bitumen matrix is compatible with the waste, and (2) measures should be taken to prevent flammability during conditioning or afterwards [23-25].

Similar recommendations as applied for vitrified waste can be used to obtain a detailed characterization of the radionuclide inventory of cemented or bitumenized waste. Of course for NPP operational waste, calculations based on the fuel are no longer relevant. Cemented and bituminized waste are known to be less homogeneous at a macro-scale (within the container), e.g. cemented ion-exchange resins. The choice of the location and amount of samples to be taken from a drum, therefore, are more difficult than for vitrified waste.

## 4.4. SIMPLE AND VARIABLE WASTE STREAMS — EXAMPLES OF NUCLEAR RESEARCH LABORATORIES

### 4.4.1. Introduction

The methodology suggested below is not applicable to production Quality Control laboratories, which are generally integrated in the production process. The subject concerns

research laboratories as well as central laboratories on nuclear sites with facilities generating waste with a very wide range of radiological characteristics.

A laboratory, whether specialized in research activities or in physical and chemical analysis, consists of several rooms. Each room includes several workstations, i.e., items installed in or connected to a containment enclosure. Typical enclosures include ventilated fume hoods, glove boxes or shielded cells.

Workstations can be dedicated to a single client for a specified time period (analysis campaign). For obvious technical and economic reasons, however, several clients use workstations. Waste collection points are often provided to centralize the waste produced in different workstations. One result of this is that waste trace ability and source segregation must be implemented by adopting a methodology that is also capable of identifying sources of error. The following discussion focuses on this aspect in particular.

In the following subsections, the term “standard reference spectrum” corresponds to the standard spectra (or fingerprint) theoretically assigned to workstations or laboratories. With regards to the term “inferred standard spectrum”, it corresponds to standard spectra previously assigned to waste collection containers from the moment they are first used, until the final examinations prior to departure from the laboratory, to the waste conditioning facility where the final waste package is produced.

#### **4.4.2. Methodology for assigning a standard spectrum to a waste container**

A standard spectrum is assigned to a waste container in several chronological steps:

— Assignment of standard reference spectra to workstations or areas

This procedure begins with the analysis of samples or wipe specimens from the workstations; then by considering the material flows entering the same workstations. Where possible, it is convenient to relate workstations assigned to a standard reference spectrum with a predominant client group having a known radiological profile (e.g. clearly defined nuclear facilities).

The stability of these relations over time makes it possible to chart the standard reference spectra of the laboratory, subject to revision at suitable intervals.

— Assignment of an inferred standard spectrum to a waste container

The inferred standard spectrum assigned to a waste container is the reference spectrum of the workstations from which the waste is collected.

In the case of a production campaign of a distinct, specific character, however, the inferred standard spectrum may correspond to the standard spectrum of the waste producer.

#### **4.4.3. General**

Whenever possible, it is advisable to specify simple and easily implemented criteria for a consistency test to limit the risk of serious errors in assigning the standard spectrum.

A typical consistency test could involve a rapid measurement of the principal  $\beta\gamma$ -emitters to check that they are present in the expected proportions. At this stage, it is important to avoid specifying unnecessarily strict and costly rules with respect to the possible impact of an error on the standard spectrum.

A deviation may be detected by the laboratory (particularly if it implements consistency testing) or by the facility in which the final waste package is conditioned. In both cases this non-quality problem must be studied. A special meeting in which all different partners compare their records with the traceability provisions must allow reassignment of the correct standard spectrum without resorting the waste drum.

#### 4.5. COMPLEX AND VARIABLE WASTE STREAMS

It is perhaps an understatement that this is the least desirable situation. The underlying objective of characterization is to first satisfy the requirements of the relevant WAC and then to balance accuracy of results with cost of analysis. The principal way of doing this is to group the wastes into streams that are sufficiently stable, so that relatively simple techniques may be applied across the population. Then as much of the properties as possible may be assigned from process knowledge from the results of an optimum number of samples. However, the complexity of these wastes means that simple techniques will not be available or suitable and the variability means that it will be unlikely that a lot of the properties will be assigned by process knowledge.

The only way to make the situation better is to find a way to partition the large complex and variable stream into identifiable smaller, more stable streams. The worst case occurs when the complex stream cannot be partitioned into different stable streams, and virtually no claims can be made about any parameter. Every waste item has to be assessed and analyzed in some detailed way. The cost and time implications of this are enormous. Conditions may involve retrieval of waste from an old store or disposal site where the wastes have been left in raw form, initial characterization has not necessarily been robust, record keeping scarce, if existing, and significant degradation of the wastes has occurred so that individual items cannot be reconciled with records (if they exist) and significant leaching and mixing has occurred.

There are essentially two ways to approach this situation, and invariably, the optimum solution depends on combining elements from each approach. Unfortunately, both are very expensive. The two approaches are:

- a. detailed analysis
- b. use of conservative limiting values

Detailed analysis is self-explanatory. Each parameter is assessed and appropriate measurements and inspections performed to build up sufficient characterization knowledge for each waste item. For example, this process could start with a sorting table or grid. Raw wastes could be spread on the table, so that each item could be visually inspected and similar materials can be grouped together for assessment. This could be followed by some sort of dose rate measurement, activity measurement for the ETM nuclides (possibly by gamma spectrometry) and fissile material assessment, if necessary. The latter, if applicable, could be carried out by NDA neutron methods or by sampling. Lastly, representative samples from

every item could be taken to determine DTM and ITM nuclide concentrations, hazardous material contamination, concentrations of corrosion enhancing materials (e.g. chlorides), complexing agents or potential gas generating substances (e.g. organics). In other words, it involves compiling the necessary information by direct observation and measurement to ensure that the final package assessment is accurate, is within limits, and the waste form complies with the WAC. To perform the above would require a very complex and impressive (and expensive) facility.

Use of limiting values is a slightly simpler approach but attracts associated penalties. This approach involves selecting a waste form and treatment process that can accommodate a wider range of parameter values. Simpler analysis approaches are then used to confirm that actual values are within the acceptable range, excluded items or compounds are not present, and a higher uncertainty in parameter values is tolerated. The types of penalties that would be encountered include the following:

- higher activities declared than are actually present and thus a higher activity disposal charge is accrued, and higher consumption of disposal site nuclide inventory than required;
- possibly the treatment process or waste form selected for each waste item is not optimal;
- process not tuned to optimum performance in order to guarantee waste form compliance leading to higher volumes of waste being disposed;
- number of rejected wastes following QC testing as uncertainties are higher.

It is obvious that either approach will be very expensive and will involve management challenges to overcome. Significant scoping research and analysis will be required to determine the best mix of techniques. A balance will have to be drawn at some point limiting the amount of up-front analysis against the benefits of downstream optimization. However, to technically defend the waste form, a significant amount of analysis will be required,

It is desirable to partition the stream into several more stable streams. As a result, the analysis burden can then be significantly reduced without a reduction in characterization quality. In most cases, partitioning will yield an increase in characterization quality along with a reduction in effort. Significant up-front scoping effort is required to achieve this result, and the aim is to strike the right balance. If partitioning is achievable, it will undoubtedly result in easier to manage streams and reduced treatment and disposal costs.

Once partitioning has occurred, the resulting waste streams will fall into one of the easier to manage categories (e.g. simple and stable, complex and stable). They can then be treated according to activities discussed in Sections 4.2 through 4.4.

Say that one partitioned stream includes the sludges. If the waste items are retrieved and sorted on a grid type tray, sludges will sift through and be collected in a tank below the table. If these sludges were well mixed prior to sampling, homogeneity could be demonstrably achieved and, in principle, only one sample would be required. Through visual identification of other items on the tray, it may be possible to associate them with a building or its original process. Not an easy task, but the benefits can be significant.

## 4.6. DECOMMISSIONING

### 4.6.1. Sampling for establishing the decommissioning strategy

Decommissioning is the process at the end of the life of a nuclear facility. The decontamination and dismantling of a nuclear facility and the cleanup of the site may produce radioactive waste with great variety in terms of type, activity, size and volume, that may be activated or contaminated.

These wastes are considered radioactive if produced by shut down nuclear facilities. Other decommissioning waste (for example accidental waste) will be addressed as historical waste. Waste streams produced by decommissioning activities are often similar to the waste produced during operation of nuclear facilities when only dry decommissioning processes are used.

If a chemical process is used (for example, chemical dissolution), there will probably be a partitioning of radionuclides, and the final radionuclide composition will be different from the initial one.

In such a situation, several sampling programmes may be necessary to determine if the radionuclide composition has changed or not, and perhaps more than one scaling factor may be necessary.

An initial sampling is often necessary to clarify the different possibilities. The main questions are as follows:

- what were all of the operations performed over the lifetime of the facility? Answering this question may require consulting archives and/or retired workers to develop a comprehensive history.
- what kind of wastes will be produced (surface or deep disposal)?
- will the waste stream be stable? If not, how many sub-waste streams will be produced, from which type (simple and stable)?
- is it technically and financially possible to decontaminate part of the waste stream to avoid producing HLW waste?
- which waste form will be produced?
- what are the required scaling factors?

Conceiving such an exhaustive characterization program is not easy. Analytical measurements are expensive and time consuming. For example, if the strategy leads to the decision to build a decontamination workshop, new information may be required by the design engineers (for safety or physical parameters). It might require some resampling. When resampling is too expensive (dose rate, time), a sampling strategy will become necessary.

### 4.6.2. Sampling strategy

Before defining a sampling strategy, it is necessary to know the complexity and history of the facility to be decommissioned. If this knowledge is not available, an NDA map of the facility may be made using a gamma camera or a gamma spectrometer.

The following sampling strategy is highly recommended when sampling is unsafe, difficult to perform, or expensive.

- define an initial sampling program taking into account the future necessity to realize complementary analytical determinations (number and weight of the samples);
- clearly identify the traceability records;
- send the samples to the laboratory in order to realize representative subsamples (necessity to clarify before the representative criteria such as KN measurements, granulometry);
- archive sufficient quantities of representative subsamples;
- process the initial analytical characterization programme to get data required to define the decommissioning strategy.

It may be possible to use aliquots of representative archived subsamples for the following needs:

- get more information relative to the waste stream to be decommissioned (special measurements required for safety, for design engineers);
- qualify the final waste form;
- determine the ratios for the DTM scaling factor.

#### 4.7. HISTORICAL WASTE

Different types of historical waste can be distinguished:

- tanks with liquids
- waste to be decontaminated before decommissioning
- workshops to be decommissioned
- waste sites (near surface, interim storage sites)

##### 4.7.1. Tanks with liquids

###### 4.7.1.1. General aspects

- There can be different subcases
  - one liquid phase
  - two different, immiscible liquid phases (organic and mineral)
  - a mixture of liquids and solids
- These radioactive wastes have to be treated before immobilization;
- The aim of the characterization here is to wisely choose the right processes considering safety and transportation.

###### 4.7.1.2. Specific recommendations for the different subcases

- One liquid phase  
The assumption of homogeneity can be confirmed based on interviews with operational personnel, knowledge of history and processes. Mixing is recommended if possible before sampling.

- Two or more immiscible phases
  1. determine the interface(s) and the volume of each phase
  2. assume each phase is homogeneous
  3. sampling has to be made in the middle of each phase
  
- Mixture of liquid and solid
 

The ratio solid/liquid should be determined, and each phase must be characterized.

  - If the tank can be mixed, sampling must be conducted during the mixing process at different depths. At each depth, a sample must be made consisting of several subsamples, one of which is to be sent to the laboratory.
    - All other subsamples are mixed, filtered and the ratio of solid to liquid and its composition are determined.
    - For all samples taken at different depths, the laboratory performs a first evaluation,
    - If chemical and physical analytical results are similar, it is assumed that the mixing process was correct and it is possible to calculate easily the S/L ratio and the composition of each.
    - If not, it is necessary to change the mixing process, requiring additional energy and time.
  
  - If it is not possible to mix the tank, then recognized specialists should study this difficult case to determine the risks and difficulties of further processing.

#### **4.7.2. Historical waste to be decontaminated before decommissioning**

##### *4.7.2.1. General aspects*

- When a chemical decontamination process is applied, one has to be aware that the initial radiochemical and chemical compositions will be modified to completely analyze the waste characteristics.
- The aim of the initial characterization is to determine the chemical, physical, and rheological properties, considering that they will significantly influence the choice of the conditioning process, to check the efficiency of the decontamination process and to determine when the clearance level is achieved. Using both NDA and DA analysis is recommended.

##### *4.7.2.2. Recommended analytical methodology*

- Measure the efficiency of the decontamination process by
  - Taking samples of the decontamination liquid waste and performing destructive analysis, and/or
  - Performing non-destructive assay
- In case of difficulties, the object (e.g. walls, counters, drains, piping) to be decontaminated should be observed with a camera and/or NDA device such as  $\gamma$ -camera to determine the specific location and type of contamination,
- Use historical records (process data sheets, chemical inventory lists, purchase records, interviews with operational personnel) and/or NDA measurements (spectrometry, neutron counting), and determine risks due to fissile materials,
- Decide for mechanical decontamination in order to reduce the final waste volume,

- Use previous data to determine the sampling plan,
- Make the chemical and radiochemical DA,
- Determine the first (chemical) decontamination process

All actions mentioned are done frequently and repeatedly.

### 4.7.3. Workshops to be decommissioned

#### 4.7.3.1. General aspects

It is assumed that in this case, the radioactive waste resulting from the decommissioning will be directly conditioned and immobilized in drums. The drums will be measured to determine the concentration of key nuclides. The aim of the characterization is to determine the scaling factors with sufficient accuracy together with the key nuclides. Checking for homogeneity and representation of the sampling and subsampling process are therefore crucial points.

#### 4.7.3.2. Recommended methodology

NDA, DA, calculation and modelling will be used. Modelling is necessary for radionuclides at or below detection limits. Three complementary steps are proposed. The first deals with measurements made in situ, the second with analytical measurements, and the third is related to the calculation and modelling step.

##### *In situ measurements*

If the historical data are accurate and well known, it will be possible to start the first two actions given here:

- Determine and locate hot spots by using a  $\gamma$ -camera, (or dose rate measurement);
- Check the consistency of the nuclide vector to decide about the location and choice of the representative active samples by using a gamma spectrometer with a counting system software;
- Take representative samples and classify them by type (metal, concrete, chemotoxic elements).

##### *Analytical measurements (for each type of sample)*

- dissolution by qualified procedures and check the representativeness of the solutions by processing standard samples of similar matrices;
- chemical separation needed prior to the measurement, depending on the measurement device (liquid scintillation, ICP-MS);
- measurement and calculation for each radionuclide of the mass and radioactivity;
- consider appropriate chemical toxicity testing.

##### *Final checking and calculation of the DTM radionuclides*

- Validation of the different DA results
  - Coherence between different samples;

- Coherence between DA results and NDA results from the in situ measurements
- Calculation of the DTM radionuclides;
  - Study the workshop history (where was the fuel processed and so on);
  - Choose a key nuclide for each hard measurable radionuclide;
  - Calculate the ratios between DTM radionuclides and the chosen KN using computer codes (ORIGEN, CESAR) on fuels processed in the workshop before decommissioning;
  - Consider process chemistry that may alter these calculated ratios and make corrections.
- Fingerprint calculation
  - Determine for each type of sample the associated fingerprint;
  - Establish the global fingerprint of the workshop by calculating it proportionally to the weight of each type of waste inside the workshop.

#### 4.7.4. Historical waste sites (near surface, interim storage sites)\*

Several questions are to be answered and information must be provided when preparing the characterization strategy for such wastes.

- What is the extent (site/volume) of the waste?
  - Determine the physical boundary of the waste site that will be retrieved and processed by physical sampling and analysis (refer to sampling standards).
  - Use any historical information and interview individuals who can provide information.
- Apply NDA to assess safety requirements for workers
- What is the condition of the waste?
  - Drummed
    - \* Intact
    - \* Breached/corroded
    - \* Solid/liquid/both
  - Contaminated equipment/supplies
  - Contaminated soil.
  - Use information to determine retrieval, transport, characterization plans
- Determine the best methods to sort the waste into similar waste streams based upon waste minimization, disposal path and anticipated waste form;
- Determine a representative sampling plan to include witness samples for archival purposes (based upon heterogeneity of waste stream). Samples should be archived until there is an approved waste conditioning process, waste form and disposal site.
- Apply detailed NDA/DA characterization of waste sorted into similar waste streams for safety, transportation and general knowledge of nuclear, chemical and physical properties.
- Based upon characterization data, determine if further processing of sorted waste streams would be beneficial for safety, waste minimization or regulatory purposes.
- Determine final waste conditioning process, waste form and disposal site.

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\*Historical waste disposal sites near the surface (trenches) or interim storage (drum waste) are considered. For very specific site cases that deal with difficult to retrieve waste, a site specific risk analysis would need to be performed to determine the waste retrieval process waste forms.

## 5. REVIEW OF CHARACTERIZATION METHODS AND TECHNIQUES

The following section provides more detailed descriptions and general guidance regarding specific characterization methodologies that are in common use. Where possible, practical guidance regarding the strengths and weaknesses of the various techniques is given. More detailed information, including details of specific methods, references, standards and reviews is provided in Annex IV.

### 5.1. NONDESTRUCTIVE EXAMINATION (NDE)

Nondestructive examination involves checking the physical and mechanical properties of the waste package. In some jurisdictions, spent ion exchange resins are not conditioned prior to storage. The design of the storage facility and substructures must be compatible with the anticipated degradation that inevitably results during long term storage.

The waste characterization strategy should include a plan for the life assessment of these storage arrangements that includes how the waste packages and storage facilities interact. Nondestructive examination of containers during the storage period should be conducted periodically. This is to assure the operators of the facility and the regulator that the waste containers are not only intact today but will remain intact until the next stage in their life cycle (e.g. retrieval and conditioning for disposal) is implemented, which could be as long as 50 to 100 years.

Typical nondestructive techniques employ wall thickness determinations using ultrasonic and holiday (porosity) detectors with high voltage discharge to determine defects in coatings. Radiography techniques are often used by waste treatment facilities to confirm that package contents conform to the WAC, e.g. to check for non-compliant items such as aerosols or free liquids or excessive void spaces.

### 5.2. NONDESTRUCTIVE ASSAY — SUMMARY OF NDA FOR WASTE CHARACTERIZATION

#### 5.2.1. General

Non-destructive assay (NDA) **involves** analysis based on observing spontaneous or stimulated nuclear radiation, interpreted to estimate the content of one or more nuclides in the item under investigation without affecting the physical or chemical form of the material. NDA of radioactive waste often involves the characterization of a waste package. Three different methods exist to perform non-destructive measurements: gamma methods, neutron methods and calorimetry. Gamma methods rely on capturing gamma photons emitted from the wastes as part of radioactive decay of the radionuclide species present; neutron methods typically rely on capture of neutrons emitted either through spontaneous fission or ( $\alpha,n$ ) reactions and are primarily used to identify and quantify fissile or fertile isotopes in the wastes; and calorimetry relies on measuring the specific heat generated from radioactive decay to quantify the amount of activity present.

NDA may be used for many different aspects of a waste characterization programme. For whom and for what purpose the information is required will largely affect what techniques are suitable. The following text provides a number of strategic issues to consider when NDA

methods are being targeted for use in characterization programmes. Brief summaries of the principal NDA methods used in various nuclear facilities are provided in the following tables. report [26]. This general guidance will be useful to help achieve the most meaningful results from NDA and will assist with development of the overall characterization plan.

A common misperception is that a waste package has not been properly characterized unless it has been subjected to NDA. What is required for proper characterization is determining an appropriate level that will be specific to the wastes, the generation process and regulatory regime. NDA is good for some applications but severely limited for others. In cases when it is not so effective, it is usually better to avoid NDA in favour of more reliable techniques (calculation or a combination of DA and calculation).

1. NDA does not by itself provide conclusive characterization. It always requires a minimum amount of process knowledge to interpret the results. The better the process knowledge, the more accurate the NDA results.
2. There has been a recent trend to develop large, complex and expensive all purpose NDA systems for use on wastes during later phases of the waste life cycle. This approach may have been desirable for waste managers because all waste can be routed through a single characterization process, thereby making the process flow chart simpler. This logic, however, is often flawed. Leaving characterization activities until later in the life cycle is giving up the opportunity to get the best quality information in the most cost effective way. Although this type of approach may be necessary to characterize historical wastes, the best approach for new wastes involves the simplest methods designed to match the given waste streams, with measurements taken as close to the time of waste generation as possible.
3. When there is an abundance of process knowledge and the characteristics are stable and reproducible, the simplest (and cheapest) NDA methods (e.g. package dose rate) may be used with a high degree of accuracy. If there is little prior knowledge or the package characteristics exhibit a high degree of variability, more sophisticated and complex (and hence, more expensive) methods must be employed, and accuracy will be severely limited. The best and most accurate results occur when the process is highly controlled, the waste characteristics are uniform, and the simplest (and least expensive) methods are fully adequate.
4. When waste characteristics are relatively uniform and a high degree of process knowledge exists, NDA may be effective with larger package sizes. When there is a high degree of variability or little specific process knowledge, accuracy may be limited by package size. This may have significant throughput considerations for a waste operations process. This may be less of a concern for a QA checking process where processing volumes and production demands may not be as severe.
5. The more sophisticated and complex the method, the higher knowledge and experience requirements for the operator. In addition, there will be increased maintenance and operational limitations—in short, the greater the potential for things to go wrong, both regarding accuracy of the measurements and in keeping the process operating. Highly complex NDA solutions are difficult to manage and generally only employed when there has not been an opportunity to gain the appropriate information in advance (e.g. historical wastes).

6. The simpler methods tend to be used for routine waste processes. More complex methods tend to be used for research and development, for quality checking laboratories, or for very complicated and difficult waste streams such as historical wastes.
7. When the highest degree of accuracy is required for each individual package, more complex methods may be required. When the accuracy of a large population is more important than that of each individual package, simpler methods may be adequate. The statistics of averaging works in our favour in this regard.
8. The best approach is to try to develop a programme whereby detailed process knowledge is obtained and preserved and the simplest, highest volume processes adequately meet accuracy requirements. The aim is to perform what is necessary and sufficient—no more and no less.

### **5.2.2. Gamma methods —A summary**

Gamma methods are effective only when a measurable amount of gamma photons are able to penetrate the package to be measured. Very low energy gamma photons (or X rays) and beta and alpha emitters cannot be detected directly by this method. However, these difficult to measure radionuclides are often scaled to an easily measured gamma emitter such as Co-60 or Cs-137. Radionuclides that decay by positron emission (e.g. Na-22) are often detected by measuring the 511 keV annihilation photons with gamma NDA instrumentation. If there is no measurable gamma emitter present in the wastes, then NDA will not be suitable as a characterization method.

Gamma methods are summarized in the tables below. Any given method will involve a combination of three aspects:

1. Measurement process
2. Type of scan
3. Matrix correction technique

Effectively, these make up a process matrix and, depending on the application, any combination is possible. Increasing complexity in any one of these aspects does not necessarily imply that increased complexity is required for the other two.

Tables V through VII provide brief descriptions of gamma NDA methods and associated matrix correction techniques.

### **5.2.3. Neutron methods —A summary**

Neutron methods are an inherently complex business. Neutron methods are used specifically to account for actinides and mostly for fissile material. Neutron methods are significantly affected by material characteristics and for wastes usually involve relatively high uncertainties. There are no simple neutron methods—each requires a knowledgeable physicist to manage the measurement campaign to ensure accurate interpretation of results.

Most mainstream methods rely on  $^3\text{He}$  proportional detector tubes. Spectroscopic methods are not in wide use nor would they typically be reliable, given that neutrons are not emitted at fixed energies. Additionally, most wastes possess a degree of moderating

properties, so all but the smallest of packages of neutron transparent materials corrupt the neutron energies well before the neutrons are captured for analysis. The source of neutrons cannot be determined simply by identifying ways they are detected. Some minimal amount of knowledge of the waste stream characteristics is required.

Neutron methods involve a combination of measurement process and matrix correction techniques. Any combination is possible. Unlike gamma measurement processes, all matrix correction techniques are inherently complex and the technique chosen needs to be closely matched to the waste stream.

Tables VIII and IX provide brief descriptions of neutron NDA methods and associated matrix correction techniques.

Table V. GAMMA MEASUREMENT PROCESS	
Dose rate	Useful for a very stable nuclide vector; it does not specifically quantify individual radionuclides. Simplest of all gamma measurements and very fast process.
Gross gamma counting	Can be very fast and makes use of large scintillation detectors (plastic or NaI). Again, useful for very stable nuclide vectors; it is not nuclide specific. Simple process. With large detectors can be very sensitive, much more than dose rate measurement.
Total spectrum	This name is a bit awkward; it is relatively the same as gross gamma counting. Spectroscopy functions can enhance gross gamma counting by using upper end discrimination to avoid naturals, visual checks of spectrum to highlight unexpected peaks, etc.
Window method or SCA	Useful when there is interference from naturals or other gamma emitters and you wish to focus on a key nuclide. Biggest advantage is speed. This does not require complex amplifiers or software, which quickly use up signal processing time.
Multiple peaks (MCA)	Specific for individual nuclides. Can independently quantify each gamma emitter. Requires more knowledgeable operator to interpret results. Beware of restricted libraries that may hide the presence of unexpected nuclides. Better to always have a warning for unidentified peaks.
HRGS vs. LRGS (CdZnTe)	Resolution determines how clearly different nuclides can be resolved from each other. Higher resolution detectors required when nuclide vectors are complex and many gammas of close energies are emitted. There is a corresponding penalty in maintenance burden and cost. LRGS detectors are more sensitive on a size for size basis and are much less expensive. Intermediate resolution detectors are showing promise, but large crystals cannot yet be grown, so process is often not applicable.
Modelling coupled with HRGS (e.g. ISOCS)	Currently being aggressively marketed as a revolutionary technique. Very complex and difficult to operate. A technique that is easy to abuse. Requires an operator with knowledge and experience equivalent of a university graduate. A technique that requires the utmost respect. It has its uses but is very difficult to manage.
Tomography	Potentially the most accurate technique; it corrects in 3 dimensions. Very powerful and no longer limited by computer processing time. Now it is limited by 3-D resolution of scan. More expensive and potentially time consuming process.
Gamma imaging (gamma camera)	Scans an area and overlays visual camera image with a survey of dose rate. Some systems incorporate spectroscopy; overlays are nuclide specific. Very powerful survey tool that helps identify where more focused surveys should be taken. Can save planning time and surveyor dose. Not usually used as final quantitative survey; uncertainties can be large when surveying a large area at a significant distance.
INCREASING COMPLEXITY <span style="float: right;">→</span>	

Spectroscopy

Table VI. TYPE OF GAMMA SCAN

INCREASING COMPLEXITY		
Total package	May be one measurement or the average of several measurements taken around the package. The total package is assumed to have average properties. May involve rotation on a turntable.	
Segmented scan	Single detector	Time consuming process; separate results are given for separate regions of the package.
	Multiple detectors	Same as for single detector but measurement time increased due to more detectors. Correspondingly more expensive.
	Vertical	Sensible to vertical variation of activity concentration.
	Angular	The Angular Scanning mode consists of collecting spectra of a waste form in a determined horizontal section (slice) at predefined angular positions. The angular scanning obtains information on the angular and/or radial non-uniformity of the distribution of activity and/or matrix properties.
	Swivelling/ horizontal	The Horizontal or Swivel scan modes merely aim at determining information on the radial distribution of activity and/or matrix properties. At a fixed height of the waste package, spectra are collected at a number of predefined horizontal positions (horizontal scanning) or at a number of predefined swivel orientations of the detector with respect to the waste form (swivel scanning). In both scanning modes (horizontal or swivel scanning) and due to the detector collimation, only a ring is visible for the detector. Depending on the stepsize used for the horizontal or swivel scan, the imaginary rings defined by the collimated detector may overlap.

Table VII. MATRIX CORRECTION TECHNIQUE FOR GAMMA METHODS		
INCREASING COMPLEXITY ↓	Operator select	Operator, through knowledge of package contents, selects appropriate calibration parameters.
	Package density	Package is weighed and fill height entered by operator; average density is computed by instrument and result corrected based on average density.
	Differential peak	Different energy peaks (e.g. Cs-134 and Cs-137) from package contents are analyzed and matrix density is computed. Requires real activity to be present in enough quantity to make computation reliable.
Active techniques	Transmission measurement	A source is shone through contents and attenuation from the other side of the package is measured. Liability in managing the source, heavy shielding required, more maintenance intensive, etc.
	Tomography	A more complex transmission measurement and thus more time consuming but much more accurate correction in 3-D without the large segment averaging uncertainties associated with SGS transmission correction.

Table VIII. NEUTRON MEASUREMENT PROCESS

	Passive total counting	Every neutron emitted is counted. Not specific to any individual actinide. Can be very sensitive due to statistical precision but easily upset by interference from other neutron emitters.
Passive Techniques	Passive coincidence counting (PNCC)	Specific to actinides that decay by spontaneous fission. If fissile material (or more appropriately, the fissionable isotopes of fissile species) is of interest, then interference from other spontaneous fission isotopes (e.g. Cf or Cm) is possible. Most often used to assess plutonium content by direct measurement of Pu-240 and inferred Pu-239 content by prior knowledge of the isotopic ratio.
	Multiplicity counting	In principle may not require separate calibrations for different material types if one of the principle parameters is accurately known. Each of the critical parameters usually varies somewhat. Potentially a powerful technique but requires careful attention.
	Active coincidence counting (ANCC)	An ever-present random neutron source induces prompt fission in fissile isotopes. Coincidence electronics rejects the random neutrons and only counts those from the fission. Useful for fissile isotopes, primarily U-235 and Pu-239.
Active Techniques	Cf-shuffler	A Cf-252 source is rapidly placed near the package and temporarily induces fission in fissile isotopes. The source is rapidly withdrawn and the delayed fission neutrons counted. The process is often repeated several times to achieve the required statistical precision. Again, useful for fissile isotopes, primarily U-235 and Pu-239. Larger packages often require excitation at several locations; the induced fission tends to be quite localized.
	Differential die-away (DDA)	A powerful neutron generator tube induces fission in fissile material with a burst of neutrons. The difference in time for the flux to decay back to normal for the package and an empty chamber determines the quantity of fissile material present. Potentially extremely sensitive. Only suitable for non-moderating materials. Very significant maintenance burden.
	Combined Passive/Active Techniques	Where wastes contain both uranium and plutonium, a combined active/passive method can be used to individually quantify each component. The passive result indicates the Pu-240 and U-238 (if present in large quantity) and the active result indicates U-235 and Pu-239 content. Mathematical combination of the results combined with knowledge of the uranium enrichment and plutonium isotopic ratio allow for computation of the separate uranium and plutonium concentrations. This is a powerful solution that is in common practice. Systems and software are more complex and require more knowledgeable users.

Table IX. MATRIX CORRECTION TECHNIQUE FOR NEUTRON METHODS	
Operator select	Each of these is too complex to summarize in this table. Very detailed descriptions and related issues are given in Section 3 of the Good Practice Guide referenced in the generic issues at the front of this summary. In general, each method suffers when neutrons can undergo significant moderation in the waste matrix prior to reaching a detector. Thus, larger packages or presence of neutron absorbers severely limit each method.
Add a source	
Flux probes	
Ring ratio	
Multiplicity	
Imaging algorithms	

### 5.3. DESTRUCTIVE ANALYSIS

Destructive analysis provides the most accurate and unbiased activity determination. Chemical and radiochemical treatment of the primary waste or waste form allows measurements to be performed that assure the traceability of the determined activity. This is not the case with NDA methods, which typically use standards calibration or modelling. As a consequence, there is greater uncertainty with NDA in terms of determining activity.

The DA process involves sampling, sample preparation and chemical separation methods.

Sampling is a critical step in the characterization process. Designing the sampling procedures and checking the homogeneity and representation of the samples assures the reliability of the final results.

Sample preparation by dissolution/mineralization has to be applied in accordance with the physicochemical characteristics of the matrix of waste forms or primary wastes and as a function of the behaviour of the specific element in the dissolution media (volatilization, precipitation). In some cases, it is possible to do a direct measurement of the radionuclide after dissolution/mineralization if the determination technique has the resolution required for it (Table X).

Chemical separation involves the chemical strategy to eliminate chemical interference (e.g. Ca in Sr determination) and radiological interference in the measurement through a chemical process such as precipitation, solvent extraction, or chromatography. (Table XI)

Radiometric determination is performed by instrumental analysis. Sophisticated methods are used such as liquid scintillation counters that allow beta spectrometry, alpha spectrometry with semiconductor detectors, high resolution gamma spectrometry for high and low energy gamma emitting nuclides, mass spectrometry that gives an accurate and efficient response for the analysis of the prepared and/or separated waste samples.(Table XII)

Table X. MINERALIZATION PROCESS

	Three step process: filtration, evaporation and dissolution in HNO <sub>3</sub>
Liquid inorganic waste	Filtration (if needed), co-precipitation and dissolution in nitric media Filtration (if needed), absorption in resins, mineralization as organic solid (see below)
Liquid organic waste	Ashing in furnace and treat as incineration ashes (see below)
Concrete	Heating acid digestion using HCl, HNO <sub>3</sub> and H <sub>2</sub> F <sub>2</sub> . The procedure is applicable in the radiochemistry determination except for halogens, compounds of C and tritium and elements that can form volatile compounds (Tc, As, Sb, Hg, B, Ru)
Bitumen	Bitumen samples are prepared in acidic media and toluene and mineralized by combustion with oxygen pressure pump. The range of application is the same as in the case of concrete samples
	Heat digestion in nitro-hydrofluoric media in the case of sludge without BaSO <sub>4</sub>
Sludge	Heat digestion in nitric media for sludge containing BaSO <sub>4</sub> . These procedures are applicable in the radiochemistry determinations except for halogens, compounds of C and tritium and elements that can form volatile compounds (Tc, As, Sb, Hg, B, Ru) By alkaline fusion with mix of metaborate of lithium and tetraborate of lithium. This method is applicable for sludge with silicates in which it is not allowed to use hydrofluoric acid and when the object of mineralization is the determination of halogens or Tc.
Incineration ashes	Alkaline fusion with Na <sub>2</sub> O <sub>2</sub> and dissolution of the product by HNO <sub>3</sub> . It is applicable to chemical and radiochemical analysis except for ashes, halogens, compounds of C and tritium and elements that can form volatile compounds (Tc, As, Sb, Hg, B, Ru)
Dissolution fines	Alkaline fusion with Na <sub>2</sub> O <sub>2</sub> and dissolution of the product by HNO <sub>3</sub> . It is applicable to chemical and radiochemical analysis of elements in the solids from dissolution of irradiated nuclear fuel except for halogens, compounds of C and tritium and elements that can form volatile compounds (Tc, As, Sb, Hg, B, Ru)
Organic products	Mineralization with H <sub>2</sub> SO <sub>4</sub> and oxidation by HNO <sub>3</sub> . Applicable to resins, vegetable and wood and to chemical and radiochemical characterization except for halogens, tritium and C compounds.

	<p>Mineralization with H<sub>2</sub>SO<sub>4</sub> and oxidation by H<sub>2</sub>O<sub>2</sub>. Applicable to ion exchange resins. Chemical and radiochemical determinations can be performed including volatile material (halogens, technetium, compounds of carbon and tritium). After mineralization another process for insoluble material treatment based on acidic attack with HCl, HNO<sub>3</sub> and H<sub>2</sub>F<sub>2</sub>, is applied for radiochemical and chemical determination of nuclides except for volatile materials.</p>
	<p>Mineralization of ion exchange resins by microwave oven with pressure control. Samples are mineralized in nitric media by heating in microwave oven with a maximum of 20 bars of pressure at 300 °C inside reactor.</p>
Soil, Filters, Combustible waste	<p>After a drying process the samples are incinerated in furnace and treated as incineration ashes.</p>

Table XI. ANALYTICAL METHODS		
NUCLIDE	SEPARATION METHODS	CHEMICAL YIELD EVALUATION
Gamma emitters	The activity determination of gamma emitters from dissolved/mineralized samples of waste can be carried out for the following nuclides using high resolution gamma spectrometry: <sup>54</sup> Mn, <sup>59</sup> Fe, <sup>58</sup> Co, <sup>60</sup> Co, <sup>65</sup> Zn, <sup>94</sup> Nb, <sup>95</sup> Nb, <sup>95</sup> Zr, <sup>106</sup> Ru, <sup>108m</sup> Ag, <sup>110m</sup> Ag, <sup>115</sup> Sn, <sup>125</sup> Sb, <sup>134</sup> Cs, <sup>137</sup> Cs, <sup>144</sup> Ce, <sup>152</sup> Eu, <sup>154</sup> Eu, <sup>155</sup> Eu.	
Tritium ( <sup>3</sup> H)	Simple or double distillation of tritiated water	
	Vacuum Distillation of tritiated water with cooling trap	
	Trapping the exhaust of microwave digestion of sample by cooling the vapour	
	Catalytic furnace to separate out <sup>3</sup> H <sub>2</sub> O/ <sup>3</sup> H <sub>2</sub> O and trapping by acidic solution; eventually further distillation processing	
Carbon-14 ( <sup>14</sup> C)	Catalytic furnace to separate out <sup>14</sup> CO <sub>2</sub> at 800°C-900°C and trapping by an amide or by alkaline solution.	
	Trapping the exhaust of microwave digestion of sample with NaOH solutions	
	Trapping of <sup>14</sup> CO <sub>2</sub> produced in the mineralization by alkaline solutions or specific scintillation cocktails with a Carbon absorber (Carbosorb®)	
	Acidolysis that is a combined process of alkaline trapping of <sup>14</sup> CO <sub>2</sub> , re-release of <sup>14</sup> CO <sub>2</sub> by acidification and purification by freezing, then mixing the frozen <sup>14</sup> CO <sub>2</sub> with Carbosorb	
Iron-55 ( <sup>55</sup> Fe)	Selective precipitation of Fe as hydroxide using NH <sub>4</sub> OH	<ul style="list-style-type: none"> <li>▪ ICP-MS</li> <li>▪ Atomic Absorption Spectrometry (AAS)</li> <li>▪ Spectrophotometry</li> </ul>
	Separation by Ion Exchange Chromatography.	

	General cationic resins or specific chromatographic columns	
	Extraction by di-isopropyl ether and selective precipitation of hydroxide in ammonia medium.	
	Direct measurement of mineralized sample	
Nickel-63 ( <sup>63</sup> Ni)	Liquid-liquid extraction of the complex <sup>63</sup> Ni – Dimethyl-glyoxime with chloroform (ClH <sub>3</sub> C)	<ul style="list-style-type: none"> <li>▪ ICP-MS</li> <li>▪ ICP-OES</li> <li>▪ AAS</li> <li>▪ Spectrophotometry</li> </ul>
	Selective precipitation as <sup>63</sup> Ni – Dimethyl-glyoxime in alcoholic media with previous purification by IEC.	
	Separation by Ion Exchange Chromatography (IEC) using specific chromatographic columns	
	Direct measurement of mineralized sample by LSC	
	Separation by Ion Exchange Chromatography (IEC) using specific chromatographic columns	
Strontium-89/Strontium-90 ( <sup>89</sup> Sr/ <sup>90</sup> Sr)	Liquid-liquid extraction with TBP and re-extraction with HNO <sub>3</sub>	<ul style="list-style-type: none"> <li>▪ ICP-MS</li> <li>▪ ICP-OES</li> <li>▪ AAS</li> </ul>
	Selective precipitation as Chromate/Carbonate	<ul style="list-style-type: none"> <li>▪ Differential weighting of purified stable Sr carrier.</li> <li>▪ γ-spectrometry of <sup>85</sup>Sr added as tracer</li> </ul>
	Selective precipitation as Oxalate/Carbonate	
	Selective precipitation as Sulphate/Carbonate	
	Separation by Ion Exchange Chromatography (IEC) using specific chromatographic columns	<ul style="list-style-type: none"> <li>▪ ICP-MS of a recovery from a known amount added (chemical analog of Tc)</li> <li>▪ LSC of a blank sample <sup>99</sup>Tc traced</li> <li>▪ γ-spectrometry of <sup>99m</sup>Tc tracer.</li> </ul>
Technetium-99 ( <sup>99</sup> Tc)	Selective extraction of TcO <sub>4</sub> <sup>-</sup> with DB18-C6 (crown ether) in toluene acetone mixture.	
	Selective extraction with tri-n-octylamine	
	Separation by precipitation	<ul style="list-style-type: none"> <li>▪ Weighting of AgI in an extra aliquot.</li> <li>▪ Measurement of <sup>129</sup>I tracer (in an aliquot or a blank).</li> </ul>
Iodine-129 ( <sup>129</sup> I)	Liquid-liquid extraction with toluene	
	Ion Exchange Chromatography (IEC) using anionic resins	

<p>Plutonium Isotopes (<math>^{238}\text{Pu}</math>, <math>^{239/40}\text{Pu}</math> &amp; <math>^{241}\text{Pu}</math>)</p>	<p>Separation by Ion Exchange Chromatography (IEC) using anionic resins washing with <math>\text{HNO}_3</math>, <math>\text{HCl}</math>, <math>\text{NH}_4\text{SCN}</math> and <math>\text{EtOH}</math>. The eluate with <math>\text{HNO}_3</math>, 8M is used for Am/Cm determination. The adsorbed plutonium is purified with <math>\text{HCl}</math>. Samples are prepared by electroplating for <math>\alpha</math>-spectrometry measurement.</p> <p>Co-precipitation of alpha emitting nuclides with ammonia using Fe as carrier. After adjustment of the valence of alpha actinides. Cm isotopes are isolated in chromatographic columns using as extractant TBP and CMPO (TRU-SPEC). Pu/Am are eluted with oxalic acid. Pu and Am are separated by IEC using AG1X4. Pu fraction is eluate with <math>\text{HCl}</math> and Am fraction is separated by elution with <math>\text{HNO}_3</math>. Samples are prepared by electroplating for <math>\alpha</math>-spectrometry measurement.</p> <p>Extraction with TOPO (in hexane). In the organic phase Pu/U isotopes are separated from other actinides. Samples are prepared by electroplating.</p> <p>Extraction of Pu/U with TPAN and elution with hexone. Samples are prepared by electroplating.</p>	<ul style="list-style-type: none"> <li>Measurement of known amounts of <math>^{236}\text{Pu}</math> or <math>^{242}\text{Pu}</math> reference standard addition.</li> <li>By difference between gross <math>\alpha</math> activity after and before extraction of Pu/U. (<math>^{241}\text{Am}</math> activity is determined by <math>\gamma</math>-spectrometry and subtracted at the gross <math>\alpha</math> determination before extraction).</li> </ul>
<p>Americium and Curium Isotopes (<math>^{241}\text{Am}</math>, <math>^{242}\text{Cm}</math> &amp; <math>^{243/44}\text{Cm}</math>)</p>	<p>Separation by Ion Exchange Chromatography (IEC) using anionic resins washing with <math>\text{HNO}_3</math>, <math>\text{HCl}</math>, <math>\text{NH}_4\text{SCN}</math> and <math>\text{EtOH}</math>. The eluate with <math>\text{HNO}_3</math>, 8M is used for Am/Cm determination. Samples are prepared by electroplating for <math>\alpha</math>-spectrometry measurement.</p> <p>Co-precipitation of alpha emitting nuclides with ammonia using Fe as carrier. After adjustment of the valence of alpha actinides. Cm isotopes are isolate in chromatographic columns using as extractant TBP and CMPO (TRU-SPEC). Pu/Am are eluated with oxalic acid. Pu and Am are separated by IEC using AG1X4. Pu fraction is eluate with <math>\text{HCl}</math> and Am fraction is separated by elution with <math>\text{HNO}_3</math>. Samples are prepared by electroplating for <math>\alpha</math>-spectrometry measurement.</p> <p>After extraction with AG1 anion exchange resin to separate Pu TRU-SPEC is used to retain U and eluate Am and Cm. Samples are prepared by electroplating for <math>\alpha</math>-spectrometry measurement.</p> <p>Extraction with TOPO (in hexane). Am/Cm remains in the aqueous phase while Pu/U isotopes are extracted in organic phase. Samples are prepared by electroplating for <math>\alpha</math>-spectrometry measurement.</p> <p>Separation by liquid-liquid extraction with TBP and Ion Exchange Chromatography (IEC) using anionic resins (Dowex 1x8). Samples are prepared by electroplating for <math>\alpha</math>-spectrometry measurement.</p> <p>Digestion and oxidation of the sample in order to prepare sample for direct measurement by ICP-MS.</p> <p>IEC using TRU-Spec (TBP-CMPO) with the residue from separation of Pu isotopes by anionic resin chromatography. The elution is electroplating in order to determine the U isotopes activity by <math>\alpha</math>-spectrometry.</p> <p>Separation of Pu by Ion Exchange Chromatography, separation of Am-Cm by IEC again and liquid-liquid extraction with MIBK. In the aqueous phase uranium isotopes can be determined.</p> <p>Extraction with TOPO (in hexane). In the organic phase Pu/U isotopes are separated from other actinides. Samples are prepared by electroplating for <math>\alpha</math>-spectrometry measurement.</p> <p>Co-precipitation with <math>\text{CeF}_4</math>.</p>	<ul style="list-style-type: none"> <li>Known amounts of <math>^{243}\text{Am}</math> reference standard are added to the sample in order to evaluate the chemical yield of the process.</li> </ul>
<p>Uranium Isotopes (<math>^{234}\text{U}</math>, <math>^{235}\text{U}</math> &amp; <math>^{238}\text{U}</math>)</p>	<p>Known amounts of <math>^{232}\text{U}</math> reference standard are added to the sample in order to evaluate the chemical yield of the process.</p>	<ul style="list-style-type: none"> <li>Known amounts of <math>^{232}\text{U}</math> reference standard are added to the sample in order to evaluate the chemical yield of the process.</li> </ul>

Table XII. ANALYTICAL METHODS

NUCLIDE	RADIOMETRIC METHODS	INTERFERENCES
Gamma emitters	The activity measurement is performed by direct measurement by high-resolution gamma spectrometry for high and low energy gamma emitting nuclides.	
Tritium ( $^3\text{H}$ )	The activity measurement is performed by Liquid Scintillation Counters (LSC) using an optimized counting window in the range 0-18.6 keV.	<ul style="list-style-type: none"> <li>▪ No special interference is described for this determination</li> </ul>
Carbon-14 ( $^{14}\text{C}$ )	The measurement for well separated $^{14}\text{C}$ is performed by LSC using a channel window in the range 4-156 keV.	<ul style="list-style-type: none"> <li>▪ <math>^3\text{H}</math>, in some cases, can contaminate carbon-isolated samples (specially in the use of catalytic furnace), which implies the use of the dual label technique (<math>^3\text{H}/^{14}\text{C}</math>) in determination of the activity. Therefore the efficiency is calculated with quenching curves established with <math>^3\text{H}</math> and <math>^{14}\text{C}</math> reference standards. The window setting optimized is 0-7.0 keV and 7.0-35.0 keV.</li> </ul>
Iron-55 ( $^{55}\text{Fe}$ )	The measurement for a separated $^{55}\text{Fe}$ is performed by LSC setting an optimized window between channels 0-8 keV. The counting efficiencies are calculated from the quenching curves performed with $^{55}\text{Fe}$ reference standard and established as a function of a quench index parameter.	<ul style="list-style-type: none"> <li>▪ In the case of direct measurement of mineralized samples, some nuclides can interfere depending on the nature of sample. <math>^{41}\text{Ca}</math>, <math>^{59}\text{Ni}</math>, <math>^{109}\text{Cd}</math> and <math>^{241}\text{Pu}</math> are described as main interferences because their maximum beta energy is in the same region as the one of <math>^{55}\text{Fe}</math> (<math>\approx 6</math> keV).</li> <li>▪ Gamma emitters interference is checked out from the sample to be measured in order to discount its contribution in the sample counting.</li> </ul>
	In the determination by low energy gamma spectrometry, efficiency is calculated through measurement in identical geometric conditions then the sample with a standard solution of $^{55}\text{Fe}$ and measurement is performed on the photopeak area at 5.8 keV.	

<p>Nickel-63 (<sup>63</sup>Ni)</p>	<p>Radioactive measurement is performed by LSC. The efficiency is calculated with quenching curves established with <sup>63</sup>Ni reference standard. The window setting optimized is 0-60 keV.</p>	<ul style="list-style-type: none"> <li>▪ The presence of other nuclides such as <sup>59</sup>Ni, <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>54</sup>Mn interfere with the measurement of <sup>63</sup>Ni by LSC.</li> <li>▪ The eventual activity of <sup>93</sup>Zr and <sup>151</sup>Sm, which are situated in nearly the same region of the β-spectrum, interfere in the case of direct measurement of mineralized samples.</li> <li>▪ Gamma emitters interference is checked out from the sample to be measured in order to discount its contribution in the sample counting.</li> </ul>
<p>Strontium-89/Strontium-90 (<sup>89</sup>Sr/<sup>90</sup>Sr)</p>	<p>LSC. The efficiency is calculated with <sup>89</sup>Sr and <sup>90</sup>Sr/<sup>90</sup>Y reference standards. The efficiency obtained is practically 100% to the different quenching values analyzed. The window setting optimized is 0-2000 keV.</p> <p>Low Level Proportional α/β detector. The measurement efficiency method is determined by using a reference solution <sup>90</sup>Sr/<sup>90</sup>Y with a specific activity.</p> <p>Geiger-Müller detector Low Level Proportional α/β detector. The measurement efficiency method is determined by using a reference solution <sup>90</sup>Sr/<sup>90</sup>Y with a specific activity.</p>	<ul style="list-style-type: none"> <li>▪ No special interferences are described</li> </ul>
<p>Tecnetium-99 (<sup>99</sup>Tc)</p>	<p>LSC. The efficiency is calculated with a <sup>99</sup>Tc reference standard.</p> <p>ICP-MS</p>	<p>Tritium is co-extracted with fraction of <sup>99</sup>Tc; however, this interference can be avoided using dual technique in the determination by LSC.</p>
<p>Iodine-129 (<sup>129</sup>I)</p>	<p>Direct measurement by ICP-MS of prepared samples</p> <p>Measurement by low energy gamma spectrometry the X ray emission of <sup>129</sup>I.</p> <p>Measurement by LSC.</p>	<p>Other radionuclides emitting gamma rays interfering with <sup>129</sup>I.</p>

<p>Plutonium Isotopes (<sup>238</sup>Pu, <sup>239/40</sup>Pu &amp; <sup>241</sup>Pu)</p>	<p><sup>241</sup>Pu is measured in separated aliquot by LSC <sup>238</sup>Pu and <sup>239/40</sup>Pu are determined by alpha spectrometry with PIPS detectors in electroplating samples</p>	<p>If good separation is performed, no major interferences are found in the final measurements. The co-precipitation of Fe in the final electroplating sample increases the self-absorption.</p>
<p>Americium and Curium Isotopes (<sup>241</sup>Am, <sup>242</sup>Cm &amp; <sup>243/44</sup>Cm)</p>	<p><sup>241</sup>Am, <sup>242</sup>Cm and <sup>242/43</sup>Cm are determined by alpha spectrometry. <sup>241</sup>Am can be determined by the gamma line of 59.5 keV.</p>	<p>Same as in the case of Pu isotopes</p>
<p>Uranium Isotopes (<sup>234</sup>U, <sup>235</sup>U &amp; <sup>238</sup>U)</p>	<p>alpha spectrometry with PIPS detectors in electroplating samples.</p>	

## 5.4. CONDITIONING PROCESS AND WASTE FORM QUALIFICATION

### 5.4.1. Introduction

Designing, testing, and building properly functioning full-scale waste conditioning facilities will be difficult if prior testing is insufficient. Scaled testing is conducted to achieve an acceptable level of risk and provide increased understanding of independent unit operations and integrated facility performance. Testing with a complete range of actual wastes to cover all process limits with regard to chemical, physical, and rheological properties requires too much actual radioactive waste that is difficult to retrieve, increases radiological safety hazards to laboratory staff, and is likely to be too time consuming and costly. Therefore, under these conditions, testing with actual radioactive waste samples is conducted to adequately characterize the initial waste, to determine the baseline properties of the conditioning process in scaled testing, and to provide a reference set of data with which to validate simulates. Simulates testing is then used to reduce the cost and hazards of developing process designs, demonstrating that the waste conditioning flowsheet is processible at the design process limits of the various unit operations, and to demonstrate process scale-up. Initial, accurate characterization of the radioactive wastes to be processed is essential.

“Simulates” are materials designed to emulate specific types of chemical or physical behaviour of actual radioactive wastes. Simulated wastes can be developed to exhibit only a limited set of important properties for a specific application or may be tailored to exhibit a broader range of chemical, physical, and rheological properties for a wide range of tests. However, proper simulant development is essential to ensure that the critical radioactive process stream characteristics are represented.

Validation of simulates is important, and the validation methodology must ensure that the necessary and sufficient actual waste properties are mimicked. Standardized chemical, physical and rheological measurement methods are necessary for measuring both the actual and simulant wastes. Use of consensus standard property test methods and reference standards when available is essential to ensure comparable results from all actual and simulant-based tests conducted.

Finally, testing of final waste forms made from actual radioactive waste is essential in demonstrating that the conditioning process and product control strategies are robust enough to produce waste forms that meet all applicable waste acceptance criteria.

### 5.4.2. Simulant development, verification and qualification

#### 5.4.2.1. *Simulant use*

The first step in simulant development is to determine the use of the simulant, e.g. studying a single precipitation, filtration, ion exchange, evaporation process or a combination of multiple unit operations. Through simulant use definition, the characteristic properties of the simulant (i.e., chemical, physical, rheological or a combination) required for development will be determined. In addition, determining what quality assurance requirements need to be met during simulant development activities is essential. Simulant development activities that support regulatory and environmental compliance related aspects of the waste conditioning process may need to meet more stringent requirements than those developed just for scoping studies.

#### 5.4.2.2. *Simulant composition defined*

Simulant compositions based upon tank samples will be defined by characterization data at the starting point. For simulant compositions that will mimic conditioned waste streams later in a waste conditioning process, flowsheet models may need to be developed (depending on the complexity of the conditioning process) to ensure the correct additional stream compositions (e.g. recycle streams from other unit operations) are addressed and incorporated. For simulant compositions formulated for specific unit operations, the composition may be tailored to only the chemical, physical, and rheological properties that are known to affect specific key operating/processing parameters. For a simulant intended to define the limits of a process or specific piece of equipment, a range of compositions should be developed to define these operational limits.

#### 5.4.2.3. *Simulant design requirement*

Developing the simulant design requirements will entail determining the necessary and sufficient simulant properties to measure (should be the same for both actual and simulant waste) for each affected unit operation, waste type, recycle stream, etc. Examples of key properties that may be critical to the conditioning process follow:

- Key processing properties. These will consist of the properties that are measured during testing of a piece of equipment or unit operation. Examples include filtrate flux rate, decontamination factors, fouling, scaling, pressure drop and sample homogeneity;
- Key chemical properties. The chemical properties of the simulant necessary to ensure a valid simulant is prepared;
- Key physical properties. Physical properties of the simulant necessary to ensure a valid simulant are prepared. Examples include density, heat capacity, thermal conductivity, heat of vaporization, particle size distribution, solids settling rate, wt% total dried solids and wt% total oxide;
- Key rheological properties. The rheological properties of the simulant would be listed. May include yield stress and shear stress versus shear rate viscosity measurements;
- Design basis range. Design assumptions employed at the particular point in the waste conditioning process would also be listed. For example, design parameters for pumps, agitators, piping and vessels that would affect the simulant will need to be documented.

Simulant developers would consult with engineers/scientists who are technically knowledgeable with all affected unit operations to determine how close each measured property must be to the target for the important analytes, physical and rheological properties. These will then become the acceptance criteria for the simulant to verify the simulant preparation procedure. Where possible, target acceptance criteria should be based upon actual waste characterization and testing data.

#### 5.4.2.4. *Develop and verify simulant meets design requirements*

Laboratory development and testing of the simulant would then be conducted to ensure the following design criteria are met during development of the simulant preparation procedure:

- Specified ionic forms of waste components were used;
- Charge balancing was completed appropriately;

- Appropriate substitutes were used for radioactive species;
- Pertinent physical properties of solids (e.g. phase, morphology, size, and crystalline vs. non-crystalline) match;
- Sequence of addition of chemical compounds or water avoids unwanted chemical reactions;
- Stipulated processing parameters important to developing a final simulant were followed.

After laboratory development and testing activities have been completed, the final simulant properties would be measured and compared with acceptance criteria. All necessary and sufficient properties should be within the acceptance criteria specifications. Note: If in the initial testing of the simulant, not all of the necessary and sufficient properties are within the acceptance criteria, it may be necessary to consult with appropriate technical personnel and possibly change the acceptance criteria. The final step to validate the simulant would be to compare testing results with similar radioactive waste testing data where available to confirm that the simulant behaviour is similar to that of actual waste.

### **5.4.3. Scaled testing facilities**

Laboratory or bench-scale testing facilities to process the actual radioactive waste will be necessary. Depending on the chemical and physical complexity of the waste streams, volume of waste to be conditioned, and number of years the waste conditioning facility will need to be in operation, an integrated pilot plant (IPP) may be necessary.

Development of an integrated pilot plant will allow integrated testing of all unit operations in an independent non-radioactive facility. The IPP would contain all necessary scaled vessels and unit operations to allow the full-scale radioactive facility to operate without affecting existing schedules. The pilot plant should be large enough to provide real engineering data but small enough to operate (e.g. cost, simulant volumes, number of operators, etc.).

The integrated pilot plant (IPP) would be designed to evaluate the entire waste conditioning envelope when all unit operations and recycle streams are interconnected. Operating the IPP would provide the following: a) data on full system operations including startup/shutdown scenarios, extended operations, and normal and off-normal processing (e.g. recognition of off-normal conditions, operator responses and appropriate recovery methods); b) optimization of the waste conditioning process flowsheet; c) confirmation of scaling data for various unit operations; d) validation of process control and compliance strategies to ensure that final waste forms meet all applicable waste acceptance criteria; and e) input to regulatory and licensing documents and agencies.

### **5.4.4. Final waste form qualification**

The primary objective for processing actual radioactive waste samples to their final waste form is to characterize the waste form produced. The actual radioactive waste samples must be processed through the same pre-treatment unit operations that will be used in the full-scale facility. Characterization of the final waste form is done to determine that all applicable regulatory waste acceptance criteria are met to allow for proper disposal. Testing of the final radioactive waste forms produced from actual radioactive waste will also show compliance with applicable contractual requirements (e.g. chemical and radionuclide reporting, waste product loading, organic content) when the waste generator, waste conditioner, and waste

disposal entities may all be managed by a separate legal identity. Regulators and independent testing verification facilities may also be required to facilitate the interaction between all parties.

## 5.5. ACCELERATED TESTING

Accelerated testing can refer to chemical, physical, mechanical, thermal and biological characteristics. An accelerated test by definition will result in increasing the rate of an alteration mode or the extent of reaction progress, when compared with expected service conditions [27]. Changes in the expected interaction mechanism(s) caused by the accelerated test conditions, if any, must be accounted for in the test data.

An overview of the different chemical, physical, mechanical, thermal and biological characteristics and the related testing was given in [1]. In practice however, accelerated testing appears to be limited to a few chemical, thermal and biological characteristics. The leach or chemical durability test is discussed below, since this is the most current characteristic tested other than the materials characteristics as measured by destructive and non-destructive analysis (see 5.2, 5.3, 5.4).

We can subdivide the chemical durability tests in two categories:

1. Tests in standard media (solutions, pH, temperature) that are used in the development stage of the waste forms or in quality assurance tests in a waste immobilization facility.
2. Tests that analyze the safety of waste management systems. Such tests currently consider the interaction of the waste form with disposal media and may involve specific conditions of pH/Eh of the solution and the presence of solids (corrosion products, backfill or disposal rock materials, radiation).

In both test categories, it is possible to apply or propose an accelerated test procedure. Acceleration is typically done by increasing the temperature (thermal activation) and/or the sample surface to solution volume (resulting in faster accumulation of dissolved species in the solution).

Tests used on low-level waste forms (cement, bitumen, polymers) are different than tests used on high-level or long-lived waste forms (glass, spent fuel, ceramics). An overview of standard tests for the two categories is given in [1].

Accelerated durability tests are particularly useful because durability testing may take a long time (months to years) before relevant data are provided. Long term data are needed when studying behaviour in geological disposal conditions. When planning accelerated durability tests, the following should be considered:

- Acceleration testing can be meaningful, especially with standard media (Category 1). It is important to verify that by increasing the temperature or the surface to volume ratio, the dissolution mechanism does not change. If it does, the acceleration effect is no longer predictable and therefore not applicable. This is of even greater concern in Category 2 tests because the waste form dissolution may become complex when it comes in contact with groundwater solutions and geological disposal media. Category 2 tests sometimes are performed at elevated temperatures (90°C and higher), although it is

- improbable that the waste form will be exposed to repository waters at such high temperatures – except in case of failure of the container.
- Category 1 tests may be used to investigate chemical durability in geological disposal conditions (Category 2 tests). This is not recommended because the dissolution processes are currently more complex in the latter situation. Category 1 tests will typically underestimate a number of phenomena such as the effects of certain solutions. For instance, the pH/Eh of groundwaters will be different from the standard solutions used in category 1 tests; category 1 tests will not reproduce the saturation effects occurring in category 2 tests and the effects of geological disposal materials. Container corrosion products and clay materials (currently used as backfill material, sometimes as repository rock) will strongly affect the waste form dissolution. It is also unrealistic to expect that an accelerated category 1 test will be able to deduce the long term dissolution rate that is observed in category 2 dissolution tests with HLW glass in pure solution.

In conclusion, it is necessary to define the objective of using accelerated tests or test conditions before starting such a programme. Knowing the objective of the test (qualification, exploitation, behaviour in normal or accidental repository conditions) will help identify the test conditions and support interpretation of the test results.

## 6. CONCLUSIONS

This publication discusses the strategy and methodology to be adopted in conceiving a characterization programme for the various kinds of radioactive waste fluxes or packages. No international publications have dealt with this topic in such depth. The strategy elaborated here takes into account the international State of the art in the different characterization methodologies.

The strategy and methodology of the characterization programme will depend on the type of radioactive waste. In addition, the accuracy and quality of the characterization programme very much depends on the requirements to demonstrate compliance with the waste acceptance criteria. This publication presents a new subdivision of radioactive waste based on its physicochemical composition and its time dependence: simple/stable, complex/stable, simple/variable and complex/variable. Decommissioning and historical waste deserve special attention in this publication, and they can belong to any of the four categories.

Identifying the life cycle of the radioactive waste is a cornerstone in defining the strategy for radioactive waste characterization. The waste acceptance criteria and the performance assessment of the repository are other key factors in the strategy and its implementation. Waste acceptance criteria may specify the inventory of specific radionuclides for the whole repository or for individual waste packages, which will affect the characterization programme. The performance assessment of the repository will draw conclusions about the critical radionuclides for disposal.

All parties involved in the generation, conditioning and disposal of the radioactive waste — including the waste generator, waste processor, characterization facility operator, repository operator and the regulator — must be involved in the conception and elaboration of the characterization strategy.

Implementation of quality control of the radioactive waste and packages by an independent laboratory, responding to the regulator, is another key issue in the strategy. The programme of this independent laboratory has to be carefully balanced with the characterization programme of the operator.

Great effort should be invested in radioactive waste characterization during the early stage of its life cycle. Characterization will be much easier at that point, and costs will be lowest. In the best overall scheme, the operator performs characterization mainly during the early phases of the life cycle, and the independent laboratory performs this activity in the later stages of the life cycle.

The scaling factor methodology is the most widely used technique to characterize radioactive waste. It is based on calculating the concentration of radionuclide “Y” from the known (by measurement or by calculation) ratio between the concentration of radionuclide “X” (called the “key” nuclide) and radionuclide “Y”. Radionuclides are further divided according to whether they are easy to measure, difficult to measure and impossible to measure. The scaling factors should be used in a proper way. Some of the recommendations are that they must be radioactive waste flux specific and must consider the chemistry of the waste. Scaling factors should be validated by destructive measures of the waste. The use of log-log plots to identify scaling factors may lead to inaccurate conclusions.

The determination and application of scaling factors will involve various kinds of non-destructive and destructive analyses of the radioactive waste. This publication discusses and recommends potential strategies for characterizing the different radioactive waste categories (simple/stable). However, there is no unique procedure that can be selected or recommended to characterize a specific radioactive waste flux. The use of non-destructive assay to measure the concentration of easy-to-measure radionuclides in the waste packages is generally recommended as a complement (at the same time as verification and updating) to the scaling factors. Upscaling information, e.g. a scaling factor, from sample level to full size drum should be done very carefully, requiring a suitable sampling strategy. The sampling of radioactive waste, in particular conditioned radioactive waste, with subsequent destructive analysis should be limited because it is expensive and difficult.

Accuracy is another cornerstone of a radioactive waste characterization programme. Accuracy is key when checking the homogeneity of a radioactive waste flux or waste package and when taking samples. Information on homogeneity and a proper choice of the samples (location, number) are needed in order to correctly transfer the data from the sample level to full size waste packages. The use of reference materials, standard procedures and comparison (proficiency) tests are strongly recommended. This publication also discusses how model calculations can be determined and how they propagate.

It is critical that the research conducted on new characterization techniques considers the objectives of the characterization of radioactive waste to include its final destination, namely, the safe disposal in surface or geological repositories.



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## Annex I

### GAMMA SCANNING

#### 1. INTRODUCTION

Due to international and national regulations for transport, interim or final storage radioactive waste has to meet specifications and criteria defined by regulatory and waste management authorities. These specifications and criteria are defined depending on the type and origin of the waste, the physical and chemical form of the waste and the type of waste package.

A **non-destructive assay (NDA)** is a procedure based on the observation of spontaneous or stimulated nuclear radiation, interpreted to estimate the content of one or more nuclides in the item which is under investigation, without affecting the physical or chemical form of the material. NDA of radioactive waste usually involves the characterization of a *waste package*.

Different non-destructive assay methods based on gamma ray spectrometry are routinely used to characterise the nuclide inventory of radioactive waste packages.

#### 2. GAMMA RAY SCANNING

##### 2.1. General

Gamma ray scanning is a procedure based on gamma ray spectrometry to measure an item in one or more sections by using one or more detectors. Segmented spectra and/or an integral spectrum are collected. Because of historic reasons ‘segmented gamma scanning’ (SGS) is often used as standard term for ‘gamma ray scanning’ to point out the importance of the segmentation process. Two types of scan modes can be described:

- **Integral gamma scanning (IGS)**, which is a procedure to measure an *integral gamma spectrum* of an item. Integral gamma ray scanning can be performed with *collimated* or *open detection geometry*.
- **Segmented gamma scanning (SGS)**: A procedure to measure one or more *segment spectra* of an item. Segmented gamma scanning requires the use of collimated *detection geometry*.

The term ‘gamma ray scanning’ is preferred because it includes both, scanning with open and collimated geometry of detection.

Gamma ray scanning can be used in all types of applications where non-destructive assay based on gamma ray spectroscopy is used for the characterization of the nuclide inventory of a volumetric radioactive source (here: radioactive waste package).

Gamma scanning is used to apply for:

- Inventory verification before waste processing or transport,
- Waste inspection before interim storage or final disposal,
- Quality checking of waste conditioning processes,
- Free release measurements.

## 2.2. Uses and field of application

Gamma ray scanning provides a non-destructive method to measure the inventory of  $\gamma$ -emitting nuclides inside a waste package where the specific nature of the matrix and the relationship between the radioactive nuclides and the matrix may be unknown.

To be representative for the complete waste package, gamma ray scanning usually involves a procedure that acquires an *integral gamma spectrum*.

Gamma ray scanning of waste packages especially involves the *correction of attenuation* effects taking place in the waste package by measurement or calibration.

Gamma ray scanning can be performed in a highly automated way with only little operator interaction. Because of the complexity of the applied equipment and procedures it shall be accomplished by quality control and quality assurance procedures.

The procedures can be applied to different types of waste packages:

- raw or unconditioned waste, including process waste (filters, resins, control rods, scrap etc.) and waste from dismantling or decommissioning,
- conditioned waste in various forms and matrices (bitumen, cement, hydraulic binder etc.),
- low level (LLW), intermediate level (ILW) and - with some restrictions - high level radioactive waste (HLW).

The containers may be of different size and shape, covering typical constructions and dimensions for shape (bag, box or cylindrical shape), shielding (possible internal shielding structures made of cast iron, concrete or lead), diameter, height, volume or mass.

Nuclides to be detected by this method must emit gamma rays with sufficient probability (i.e., intensity and energy) to penetrate the surrounding matrix material. The useful energy range is dependent on the type and composition of the matrix, the source position or source distribution inside the package and the type and dimension of the container.

## 2.3. Methods

There are many different types of systems in use to perform gamma ray scanning of waste packages. Gamma ray scanning is performed by an instrument, usually called a *gamma ray scanner*. It is accomplished by a method which enables the operator to calculate the activity of nuclides of interest from the recorded spectrum or a set of recorded spectra of gamma emitters contained in the waste package.

State of the art systems consist of a gamma-spectrometry system, an appropriate mechanical device to perform and control the movement of the detector(s) or waste package and a control and data processing unit. In general gamma scanners use a turntable to rotate the waste package during the scanning process. Rotation is primarily performed to average radial and angular variations of the system response for non-homogenous waste.

Two different system configurations are distinguished according to the detection geometry and type of scan procedure:

- Open detection geometry to perform integral gamma scanning (IGS, refer to fig. 2),
- Collimated detection geometry “segmented gamma scanning” (SGS, refer to fig. 3) or “integral gamma scanning” (which is the term used preferentially by the US NRC).

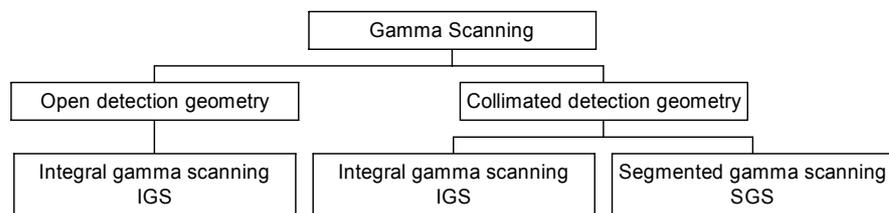


FIG. 1. Basic classification of gamma scanning

IGS can be performed with both open and collimated detection geometry and is mainly performed to acquire an integral spectrum of the waste package.

In the segmented mode the procedure can also provide *vertical, horizontal* and/or *angular profiles* of the specific gamma ray emission of nuclides of interest from the counts or count rates distributions measured at the different scanning positions. These profiles can help to characterise the activity distribution inside a waste package.

#### 2.4. Open detection geometry (see Fig. 2)

The basic configuration for this type of gamma ray scanning involves only one detector in a fixed position relative to the waste package. The open geometry configuration is set-up in such a way that all parts of a package can contribute to the response of the detector (integral mode).

Systems based on the open geometry procedure have the advantage of less complicated hardware and generally higher detection efficiency compared to systems applying a collimated geometry and segmented mode of scanning. In comparison, open geometry measurements usually lead to significantly lower detection thresholds. However, this method is generally more sensitive to the distribution of activity and density of the waste matrix.

Open geometry measurements are typically applied for NDA of low level waste (LLW) packages with homogeneous or well-characterised filling.

Calibration is usually performed by using reference packages containing certified reference material (calibration sources) or analytical calculations.

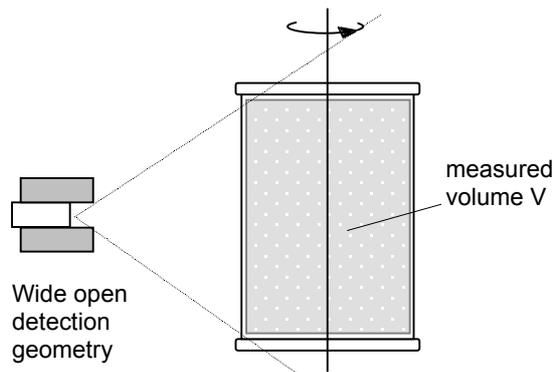


FIG. 2. Sketch of the open geometry mode of gamma ray scanning

## 2.5. Collimated detection geometry (see Fig. 3)

For intermediate level waste (ILW) or high level waste (HLW) and in order to operate at acceptable dead-times of the detection system, the following parameters can be optimised:

- Reduction of the measurement volume by appropriated choice of collimator aperture and/or the distance between detector and waste package. In this way the system is able to cope with various levels of gamma flux (counting rates) emitted by the waste package.

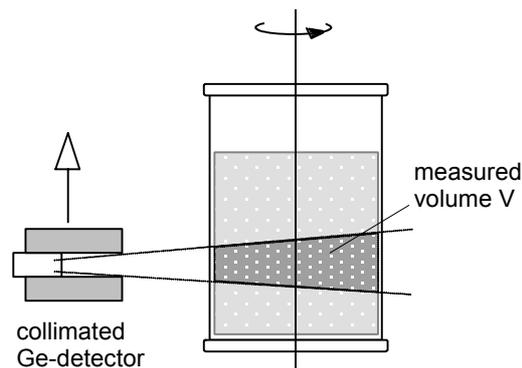


FIG. 3. Sketch of the segmented mode of gamma ray scanning

The collimated detection geometry is primarily used to perform SGS, i.e., to record segment spectra which are representative for specific volume parts of a waste package. Segmentation may refer to:

- vertical (vertical scanning see fig. 4a),
- horizontal (horizontal scanning see fig. 4b) or
- angular subdivisions (angular scanning see fig. 4c).

The acquisition of segment spectra is performed at a predefined set of positions of the detection system close to the surface of a waste package. In SGS an integral spectrum of the total nuclide inventory is determined from a set of segment spectra being representative for the complete waste package.

In general, and in combination with a segment-specific determination of *attenuation factors* (i.e., transmission measurement), SGS offers the possibility of segment-specific attenuation correction. This usually results in being less sensitive to non-uniform activity and density distributions.

Segment specific spectra can be used to check the adherence of the waste package to underlying assumptions for activity calculation or calibration such as uniformity of source and matrix distribution.

This facilitates the definition and evaluation of criteria for conformity checking, which is especially important for waste forms with 'unknown' specification.

Calibration is generally performed by using reference packages containing certified reference material (calibration sources) or analytical calculations.

## **2.6. Additional characteristics**

Both types of gamma ray scanning may involve the use of more than one detector to increase system throughput.

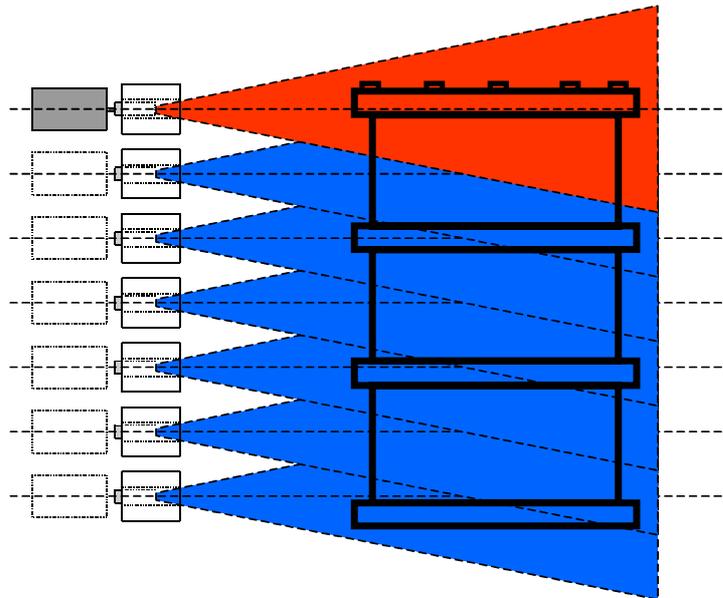
State of the art SGS systems apply techniques to determine the attenuation properties of the matrix experienced by the radiation emitted from nuclides inside the waste package. Attenuation correction can be performed by one of the following methods:

— *weight correction*

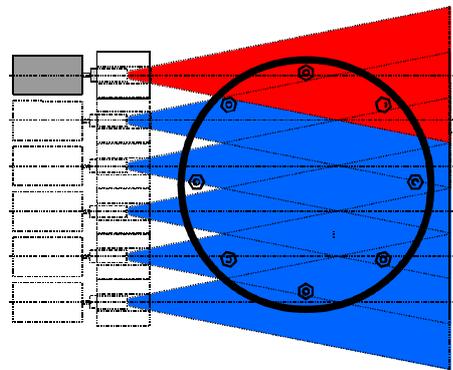
This involves the determination of the apparent density of the waste matrix which is a fundamental technique in SGS because the weight can always be determined:

- *Differential peak absorption technique (DPA)*, which may be applied if gamma rays with different energies can be detected from the same nuclide,
- *Evaluation of scattered gamma rays*, the technique is based on a detailed evaluation of the pulse height spectrum to assess the 'amount' of scattered gamma rays.
- *Transmission corrected gamma ray scanning* this implies the use of an external gamma ray source to measure the transmission at one or more energies for a segment,
- *Transmission computed tomography (TCT)*, yields a two-dimensional distribution of attenuation factors within a (usually) horizontal slice of the waste package.

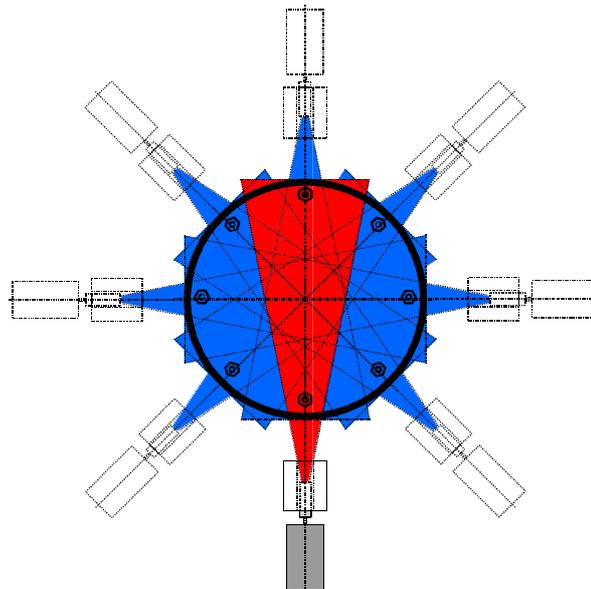
Segmented gamma ray scanning can be extended in such a way that low-resolution tomography may be performed.



**Fig. 4A) VERTICAL SCANNING**



**Fig. 4B) HORIZONTAL SCANNING**



**Fig. 4C) ANGULAR SCANNING**

*FIG. 4. Sketches of different conventional scanning modes used in segmented gamma ray scanning (vertical, horizontal and angular scanning).*

**Transmission computed tomography (TCT):** A NDA method which allows the distribution of matrix density to be determined within sections of waste package. The technique is based upon the detection of gamma rays emitted from a known source and the calculation of transmission ratios through specific paths within the waste matrix

**Emission computed tomography (ECT):** A NDA method which allows the distribution of nuclide activity to be determined within sections of waste package. The technique is based upon the measurement of segmented spectra from known sections of the waste matrix in order to obtain accurate results it is necessary to know the distribution of matrix density within the section (typically by TCT).



## Annex II

### LIST OF RADIONUCLIDES

The following are the principal radionuclides likely to be found in the initial reprocessing sludge in a spent fuel reprocessing facility:

— Actinides

- $^{244}\text{Cm}$ ,  $^{243}\text{Cm}$ ,  $^{242}\text{Cm}$ ;
- $^{243}\text{Am}$ ,  $^{242}\text{Am}$ ,  $^{241}\text{Am}$ ;
- $^{237}\text{Np}$ ;
- $^{238}\text{U}$ ,  $^{236}\text{U}$ ,  $^{235}\text{U}$ ,  $^{234}\text{U}$ ,  $^{233}\text{U}$ ,  $^{232}\text{U}$ ;
- $^{242}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{238}\text{Pu}$ ;
- $^{233}\text{Pa}$ ,  $^{231}\text{Pa}$ ;
- $^{234}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{228}\text{Th}$ .

— Fission products

- $^3\text{H}$ ,  $^{79}\text{Se}$ ,  $^{85}\text{Kr}$ ,  $^{90}(\text{Sr}+\text{Y})$ ,  $^{93}\text{Zr}^*$ ,  $^{95}(\text{Zr}+\text{Nb})^*$ ,  $^{99}\text{Tc}$ ,  $^{103}\text{Ru}$ ,  $^{106}(\text{Ru}+\text{Rh})$ ,  $^{129}\text{I}$ ,  $^{134}\text{Cs}$ ,  $^{135}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{144}(\text{Ce}+\text{Pr})$ ,  $^{147}\text{Pm}$ ,  $^{151}\text{Sm}$ ,  $^{154}\text{Eu}$ ,  $^{155}\text{Eu}$ .  
 *$^{93}\text{Zr}$  and  $^{95}(\text{Zr}+\text{Nb})$  are also activation products.*

— Structural component activation products; their formation depends on the quality of the materials used (alloy grades, nature and impurity content) and subjected to irradiation. The following groups with similar chemical behavior may be distinguished within these radionuclides:

- $^{36}\text{Fe}$ ,  $^{57}\text{Co}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$ . The most suitable tracer is  $^{60}\text{Co}$  because of its abundance and radioactive half-life. The impurity concentrations in the structural materials may result in significant variations in the relative activities of these radionuclides.
- $^{107}\text{Pd}$ ,  $^{106}(\text{Ru}+\text{Rh})$ ,  $^{103}\text{Ru}$ : these are platinum-group metals with similar chemical behavior. The radioactive half-life of  $^{106}\text{Ru}$  makes it a better tracer than  $^{103}\text{Ru}$ , but it is poorly suitable for extended cooling times;
- $^{144}\text{Ce}$ ,  $^{147}\text{Pm}$ ,  $^{151}\text{Sm}$ ,  $^{154}\text{Eu}$ ,  $^{155}\text{Eu}$ : these are lanthanides with comparable chemical behavior. The radioelements  $^{144}\text{Ce}$ ,  $^{154}\text{Eu}$  and  $^{155}\text{Eu}$  are directly measurable by  $\gamma$  spectrometry, but the half-life and  $\gamma$  spectrum emitted by  $^{154}\text{Eu}$  make it a better tracer;
- $^{134}\text{Cs}$ ,  $^{135}\text{Cs}$ ,  $^{137}\text{Cs}$ : these are all isotopes of the same chemical element. Because of its radioactive half-life and rate of formation in the reactor,  $^{137}\text{Cs}$  is the best tracer of  $^{135}\text{Cs}$ ;
- $^{244}\text{Cm}$ ,  $^{243}\text{Cm}$ ,  $^{242}\text{Cm}$ ,  $^{241}\text{Am}$ ,  $^{242\text{m}}\text{Am}$ ,  $^{243}\text{Am}$ : these trivalent actinides exhibit similar chemical behavior during spent fuel reprocessing.  $^{241}\text{Am}$  is “directly measurable” by  $\gamma$  spectrometry, provided the 59.6 keV  $\gamma$  line characteristic of this radionuclide is measured under carefully controlled conditions.



**Annex III**  
**RELEVANT STANDARDS**

*Remark: the list given below does not pretend to be complete.*

<i>ISO STANDARDS</i>
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*ISO Technical Committee 85 on Nuclear Energy has a subcommittee (SC5) relative to fuel cycle. Within SC5, Working group 5 deals with characterization of radioactive wastes*

**Project 14850 part 3**

Waste-packages activity measurement – high resolution gamma spectrometry using segmented gamma scanning the first draft has been produced by ENTRAP.

**NWIP N701** (NWIP = New work item proposal)

Standard guide for the scaling factor method to determine the radioactivity of low and intermediate level radioactive waste packages generated at nuclear power plants.

— **ISO 6962** (*published in June 2004*)

Standard method for testing the long-term alpha irradiation stability of matrices for solidification of high-level radioactive waste.

— **FDIS ISO 16797** (FDIS = Final draft international standard)

Soxhlet-mode chemical durability test – Application to vitrified matrixes for high-level radioactive waste.

— **DIS ISO 14850 part 1** (DIS = Draft international standard)

Standard guide for gamma ray spectrometric measurements of radioactive waste packages using HPGe-detectors.

— **FDIS 14850 part 2**

Waste-packages activity measurement – high resolution gamma spectrometry in integral mode with open geometry.

<i>ASTM STANDARDS (USA)</i>
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American National Standards Institute, Inc., American Nuclear Society, ANSI/ANS - 16.1 - 1986, “American National Standard Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure”, La Grange Park, Illinois.

American Nuclear Society, “Design Guides for Radioactive Material Handling Facilities and Equipment” ISBN 0-89448-554-7 American Nuclear Society, La Grange Park, Illinois 60525, 1988

ASTM International C998-90 (Reapproved 2000) “Standard Practice for Sampling Surface Soil for Radionuclides”, West Conshohocken, Pennsylvania.

ASTM International C982-88 (Reapproved 1997) “Standard Guide for Selecting Components for Energy-Dispersive X ray Fluorescence (XRF) Systems”, West Conshohoken, Pennsylvania.

ASTM International C998-90 (Reapproved 2000) “Standard Practice for Sampling Surface Soil for Radionuclides”, West Conshohoken, Pennsylvania.

ASTM International C999-90 (Reapproved 2000) “Standard Practice for Soil Sample Preparation for the Determination of Radionuclides”, West Conshohoken, Pennsylvania.

ASTM International C1000-00 “Standard Test Method for Radiochemical Determination of Uranium Isotopes in Soil by Alpha Spectrometry”, West Conshohoken, Pennsylvania.

ASTM International C1001-00 “Standard Test Method for Radiochemical Determination of Plutonium in Soil by Alpha Spectroscopy”, West Conshohoken, Pennsylvania.

ASTM International C1030-95 (Reapproved 2001) “Standard Test Method for Determination of Plutonium Isotopic Composition by Gamma ray Spectrometry”, West Conshohoken, Pennsylvania.

ASTM International C1062-00 “Standard Guide for Design, Fabrication, and Installation of Nuclear Fuel Dissolution Facilities”, West Conshohoken, Pennsylvania.

ASTM International C1109-98 “Standard Test Method for Analysis of Aqueous Leachates from Nuclear Waste Materials Using Inductively Coupled Plasma-Atomic Emission Spectrometry”, West Conshohoken, Pennsylvania.

ASTM International C1111-98 “Standard Test Method for Determining Elements in Waste Streams by Inductively Coupled Plasma-Atomic Emission Spectroscopy”, West Conshohoken, Pennsylvania.

ASTM International C1133-96 “Standard Test Method for Nondestructive Assay of Special Nuclear Material in Low Density Scrap and Waste by Segmented Passive Gamma ray Scanning”, West Conshohoken, Pennsylvania.

ASTM International C1174-97 “Standard Practice for Prediction of the Long-Term Behavior of Materials, Including Waste Forms, Used in Engineered Barrier Systems (EBS) for Geological Disposal of High-Level Radioactive Waste”, West Conshohoken, Pennsylvania.

ASTM International C1205-97 (Reapproved 2002) “Standard Test Method for The Radiochemical Determination of Americium-241 in Soil by Alpha Spectrometry”, West Conshohoken, Pennsylvania.

ASTM International C1207-97 “Standard Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting”, West Conshohoken, Pennsylvania.

ASTM International C1217-00 “Standard Guide for Design of Equipment for Processing Nuclear and Radioactive Materials”, West Conshohoken, Pennsylvania.

ASTM International C1220-98 “Standard Test Method for Static Leaching of Monolithic Waste Forms for Disposal of Radioactive Waste”, West Conshohoken, Pennsylvania.

ASTM International C1221-92 (Reapproved 1998) “Standard Test Method for Nondestructive Analysis of Special Nuclear Materials in Homogeneous Solutions by Gamma ray Spectrometry”, West Conshohoken, Pennsylvania.

ASTM International C1234-98 “Standard Practice for Preparation of Oils and Oily Waste Samples by High-Pressure, High-Temperature Digestion for Trace Element Determinations”, West Conshohoken, Pennsylvania.

ASTM International C1255-93 (Reapproved 1999) “Standard Test Method for Analysis of Uranium and Thorium in Soils by Energy Dispersive X ray Fluorescence Spectroscopy”, West Conshohoken, Pennsylvania.

ASTM International C1268-94 (Reapproved 2000) “Standard Test Method for Quantitative Determination of Americium 241 in Plutonium by Gamma ray Spectrometry”, West Conshohoken, Pennsylvania.

ASTM International C1284-00 “Standard Practice for Electrodeposition of the Actinides for Alpha Spectrometry”, West Conshohoken, Pennsylvania.

ASTM International C1285-02 " Standard Test Methods for Determining Chemical Durability of Nuclear, Hazardous, and Mixed Waste Glasses and Multiphase Glass Ceramics: The Product Consistency Test (PCT)", West Conshohoken, Pennsylvania.

ASTM International C1308-95 (2001) “Standard Test Method for Accelerated Leach Test for Diffusive Releases from Solidified Waste and a Computer Program to Model Diffusive, Fractional Leaching from Cylindrical Waste Forms”, West Conshohoken, Pennsylvania.

ASTM International C1310-01 “Standard Test Method for Determining Radionuclides in Soils by Inductively Coupled Plasma-Mass Spectrometry Using Flow Injection Preconcentration”, West Conshohoken, Pennsylvania.

ASTM International C1316-01 “Standard Test Method for Nondestructive Assay of Nuclear Material in Scrap and Waste by Passive-Active Neutron Counting Using a <sup>252</sup>Cf Shuffler”, West Conshohoken, Pennsylvania.

ASTM International C1343-96 (Reapproved 2002) “Standard Test Method for Determination of Low Concentrations of Uranium in Oils and Organic Liquids by X ray Fluorescence”, West Conshohoken, Pennsylvania.

ASTM International C1387-98 “Standard Guide for Determination of Technetium-99 in Soil”, West Conshohoken, Pennsylvania.

ASTM International C1402-98 “Standard Guide for High-Resolution Gamma ray Spectrometry of Soil Samples”, West Conshohoken, Pennsylvania.

ASTM International C1416-99 “Standard Test Method for Uranium Analysis in Natural and Waste Water by X ray Fluorescence”, West Conshohoken, Pennsylvania.

ASTM International C1455-00 “Standard Guide for Nondestructive Assay of Special Nuclear Material Holdup Using Gamma ray Spectroscopic Methods”, West Conshohoken, Pennsylvania.

ASTM International C1458-00 “Standard Test Method for Nondestructive Assay of Plutonium, Tritium and <sup>241</sup>Am by Calorimetric Assay”, West Conshohoken, Pennsylvania.

ASTM International C1463-00 “Standard Practices for Dissolving Glass Containing Radioactive and Mixed Waste for Chemical and Radiochemical Analysis”, West Conshohoken, Pennsylvania.

ASTM International C1475-00 “Standard Guide for Determination of Neptunium-237 in Soil”, West Conshohoken, Pennsylvania.

ASTM International C1490-01 “Standard Guide for the Selection, Training and Qualification of Nondestructive Assay (NDA) Personnel”, West Conshohoken, Pennsylvania.

ASTM International C1493-01 “Standard Test Method for Non-Destructive Assay of Nuclear Material in Waste by Passive and Active Neutron Counting Using a Differential Die-Away System”, West Conshohoken, Pennsylvania.

ASTM International C1500-02 “Standard Test Method for Nondestructive Assay of Plutonium by Passive Neutron Multiplicity Counting”, West Conshohoken, Pennsylvania.

ASTM International C1507-01 “Standard Test Method for Radiochemical Determination of Strontium-90 in Soil”, West Conshohoken, Pennsylvania.

ASTM International C1514-02 “Standard Test Method for Measurement of <sup>235</sup>U Fraction using the Enrichment Meter Principle”, West Conshohoken, Pennsylvania.

ASTM International C1533-02 “Standard Guide for General Design Considerations for Hot Cell Equipment”, West Conshohoken, Pennsylvania.

ASTM C 1174-97. 1997, Standard Practice for Prediction of the Long-Term Behavior of Materials, Including Waste Forms, Used in Engineered Barrier Systems

<i>CETAMA and AFNOR (France)</i>
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## Non-destructive analysis (NDA)

No.	AFNOR standard	CETAMA contribution
302	<p><a href="#">NF M60-302</a> (January 2000)  <b>Classification:</b> M60-302  <b>ICS code(s):</b> 13.030.30, 13.280  <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Measurement of the activity of waste packages by high resolution gamma spectrometry  <b>Number of pages:</b> 15  <b>Supersedes:</b> NF M60-302:199612 (M60-302)</p>	WG 27
303	<p><a href="#">NF M60-303</a> (April 1999)  <b>Classification:</b> M60-303  <b>ICS code(s):</b> 13.030.30, 17.240  <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Recommendations for calibration of a gamma spectrometry measuring system for radioactive waste package activity.  <b>Number of pages:</b> 13</p>	WG 27
305	<p><a href="#">NF M60-305</a> (April 1999)  <b>Classification:</b> M60-305  <b>ICS code(s):</b> 13.030.30, 17.240  <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Radionuclide inventory by nondestructive measurement of radionuclide tracers  <b>Number of pages:</b> 9</p>	
306	<p><a href="#">NF M60-306</a> (October 1998)  <b>Classification:</b> M60-306  <b>ICS code(s):</b> 13.030.30, 17.240  <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Determination of the radioactive characteristics of waste packages by passive neutron counting  <b>Number of pages:</b> 25</p>	WG 27
309	<p><a href="#">NF M60-309</a> (April 2001)  <b>Classification:</b> M60-309  <b>ICS code(s):</b> 13.030.30, 17.240  <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Measurement of tritium gas release from tritiated waste packages using an ionization chamber  <b>Number of pages:</b> 12</p>	–
315	<p><a href="#">NF M60-315</a> (March 2001)  <b>Classification:</b> M60-315  <b>ICS code(s):</b> 13.030.30, 17.240  <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Recommendations for calibration of a measurement station to determine the radioactive characteristics of waste packages by passive neutron counting  <b>Number of pages:</b> 44</p>	WG 27

## Destructive analysis (DA)

No.	AFNOR standard	CETAMA contribution
313	<p><a href="#">NF M60-313</a> (October 1998)  <b>Classification:</b> M60-313  <b>ICS code(s):</b> 13.030.30, 17.240  <b>Title:</b> Nuclear energy – Nuclear fuel cycle technology. Waste – Soxhlet mode test of chemical durability. Application to materials arising from vitrification processes.  <b>Number of pages:</b> 12</p>	–

No.	AFNOR standard	CETAMA contribution
316	<a href="#">NF M60-316</a> (May 2002) <b>Classification:</b> M60-316 <b>ICS code(s):</b> 13.030.30, 17.240 <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Determination of strontium-90 in liquid effluents and solid waste after prior chemical separation <b>Number of pages:</b> 21	WG 14 Method 375
317	<a href="#">NF M60-317</a> (March 2001) <b>Classification:</b> M60-317 <b>ICS code(s):</b> 13.030.30 <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Determination of nickel-63 in liquid effluents and solid waste by liquid scintillation, after prior chemical separation <b>Number of pages:</b> 15	WG 14 Method 374
318	<a href="#">NF M60-318</a> (April 2002) <b>Classification:</b> M60-318 <b>ICS code(s):</b> 13.030.30, 17.240 <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Characterization of the thermal reactivity of bituminized materials by microcalorimetry <b>Number of pages:</b> 18	–
320	<a href="#">NF M60-320</a> (February 2004) <b>Classification:</b> M60-320 <b>ICS code(s):</b> 13.030.30, 17.240, 27.120.30 <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Determination of the carbon-14 activity in liquid effluents and solid waste by liquid scintillation <b>Number of pages:</b> 17	WG 14 Method 377
322	<a href="#">NF M60-320</a> (to be published) <b>Classification:</b> M60-322 <b>ICS code(s):</b> <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Determination of the iron-55 activity in liquid effluents and solid waste by liquid scintillation after prior chemical separation <b>Number of pages:</b>	WG 14 Method 379
325	<a href="#">NF M60-320</a> (to be published) <b>Classification:</b> M60-325 <b>ICS code(s):</b> <b>Title:</b> Nuclear energy – Fuel cycle technology. Waste – Guidelines for dissolution prior to analysis of liquid effluents, solid waste and encapsulation matrices <b>Number of pages:</b>	WG 14 Dissolution guidelines

#### *OTHER STANDARDS*

- S.C. Goheen, M. McCulloch, B.L. Thomas, R.G. Riley, D.S. Sklarew, G.M. Mong, and S.K. Fadeff, “DOE Methods for Evaluating Environmental and Waste Management Samples,” Publication number DOE/EM-0089T, Rev. 2, Pacific Northwest National Laboratory, Richland, WA 99352, October 1994.
- SW-846, Test Methods for Evaluating Solid Waste, Physical/Chemical Methods, U.S. Environmental Protection Agency, Office of Solid Waste and Emergency Response, Washington, D.C.
- Toxicity Characteristic Leaching Procedure (TCLP). 1992. SW-846, Method 1311. Rev. 2. In Test Methods for Evaluating Solid Waste, Volume 1C: Laboratory Manual Physical/Chemical Methods, U.S. Environmental Protection Agency, Office of Solid Waste and Emergency Response, Washington, D.C.

## Consensus standards annotated bibliography

### American National Standards Institute (ANSI) Standards Institute of Nuclear Materials Management

- ANSI Standards under the Jurisdiction of N-15 Technical Committee on Methods of Nuclear Material Control
- ANSI N15.41 R(1994) “Nuclear Facilities – Derivation of Measurement Control Programs – General Principles”

This standard presents general principles from which specific measurement control programs can be derived. The purpose of the standard is to provide the principles for deriving a sound measurement control program for any measurement process used to manage or to safeguard nuclear materials. It is the overarching “umbrella standard” defining basic requirements of any measurement control program; other standards (N15.36, N15.19, N15.51) offer more specific information about measurement control programs for nondestructive assay, volume tank calibration, and analytical chemistry laboratories, respectively. Although the principles presented in this standard are intended primarily to be applied to the measurement processes used in the nuclear industry, the principles are general enough for application to many other measurement processes as well.

- ANSI N15.51 R(1996) “Nuclear Materials Management – Measurement Control Program – Nuclear Materials Analytical Chemistry Laboratory”

This standard is based on N15.41. It provides the principle elements of a measurement control program for an analytical chemistry laboratory supporting nuclear fuel cycle activities, nuclear material safeguards, accounting, and process quality. The goal of any measurement control program is to publication and quantify the performance of each analytical measurement system and to provide for detection and correction of adverse changes. For those facilities that must conduct physical inventories, the measurement control program provides data for establishing the uncertainty (or limits of error) associated with a given inventory difference value.

- ANSI N15.36 (1994) “Nuclear Materials – Nondestructive Assay Measurement Control and Assurance”

This standard is based on N15.41. It provides the principle elements of a measurement control program for nondestructive assay measurements of nuclear material by observation of spontaneous or stimulated radioactive emissions. The guidelines given in this standard for the establishment and operation of an NDA measurement control program may be appropriate for other types of measurements; the monitoring and control techniques recommended are designed specifically for NDA radioactive emissions measurement situations and may not be applicable in other measurement methods.

**Other potentially applicable N-15 Technical Committee on Methods of Nuclear Material Control standards include:**

- N15.28 “Guide for Qualification and Certification of Safeguards and Security Personnel”
- N15.19 “Nuclear Materials Control – Volume Calibration Techniques” (under revision)
- N15.10 “Nuclear Materials – Unirradiated Plutonium Scrap – Classification”

American Nuclear Society Standards

- ANSI/ANS-16.1-2003 “Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure”

This standard, ANSI/ANS-16.1-2003, provides a uniform procedure to measure and index the release of radionuclides from waste forms as a result of leaching in demineralized water for 5 days. The results of this procedure do not apply to any specific environmental situation except through correlative studies of actual disposal site conditions. The test presented in this standard has much in common with the original International Atomic Energy Agency proposal and has by now become familiar to those working in the radioactive waste-form development field. It contains the provisions published in the original version of this standard in 1986.

- ANSI/ANS-55.1-1992;R2000 -- “Solid Radioactive Waste Processing System for Light-Water-Cooled Reactor Plants”

This standard sets forth the design, construction, and performance requirements for a solid radioactive waste processing system for light-water-cooled reactor plants. For the purposes of this standard, the solid radioactive waste processing system begins at the interface with the liquid radioactive waste processing system boundary and at the inlets to the spent resin, filter sludge, evaporator concentrate, and phase separator tanks. In addition, this standard pertains to dry active waste, mixed waste, and other solid radioactive waste forms that are generated as part of the operation and maintenance of light-water-cooled reactor plants. The system includes facilities for temporary (up to 30 days of anticipated normal waste generation) on-site storage of packaged waste but terminates at the point of loading the filled drums and other containers on a vehicle for shipping off-site to a licensed disposal site or transfer to interim (up to 5 yr.) on-site storage facilities. The solid radioactive waste processing system is not a safety-class system as defined by American National Standard Nuclear Safety Criteria for the Design of Stationary Pressurized Water Reactor Plants, ANSI/ANS-51.1-1983 (R1988) or as defined in American National Standard Nuclear Safety Criteria for the Design of Stationary Boiling Water Reactor Plants, ANSI/ANS-52.1-1983 (R1988).

- ANS-55.4-1993;R1999 – “Gaseous Radioactive Waste Processing Systems for Light Water Reactor Plants”

This standard sets forth minimum design, construction, and performance requirements, with due consideration for operation, for gaseous radioactive waste processing systems (GRWPS) for light water reactor (LWR) plants. It is applicable for routine operation, design basis fuel leakage, and other design basis occurrences.

- ANS-55.6-1993;R1999-- Liquid Radioactive Waste Processing System for Light Water Reactor Plants

This standard sets forth minimum design, construction, and performance requirements, with due consideration for operation, of the Liquid Radioactive Waste Processing System (LRWPS) for light water reactor (LWR) plants for design basis inputs. It is applicable to routine operation, including design basis fuel leakage and other design basis occurrences.

- “Design Guides for Radioactive Material Handling Facilities and Equipment” American Nuclear Society, 1988

The guide provides general requirements for design, construction, operation, maintenance, and decommissioning of facilities dealing with radioactive materials and specifically remote handling facilities. The guide covers remote facilities such as fusion experiments accelerators, nuclear fuel handling and examination and reprocessing facilities, remote fuel fabrication facilities, and related nuclear hot cells. The guide does not cover areas related to glove boxes, mining, radioisotope laboratories, radiopharmaceuticals, and reactors.

#### ASTM International Standards

##### **ASTM Standards under the Jurisdiction of C26.05 Methods of Test**

- C982-03 “Standard Guide for Selecting Components for Energy-Dispersive X ray Fluorescence (XRF) Systems”

This guide describes the components for an energy-dispersive X ray fluorescence (XRF) system for materials analysis. It can be used as a reference in the apparatus section of test methods for energy-dispersive X ray fluorescence analyzes of nuclear materials.

The components recommended include X ray detectors, signal processing electronics, data acquisition and analysis systems, and excitation sources that emit photons.

Detailed data analysis methods are not described or recommended, as they may be unique to a particular analysis problem. Some applications may require the use of spectrum deconvolution to separate partially resolved peaks or to correct for matrix effects in data reduction.

- C998-90 (Reapproved 2000) “Standard Practice for Sampling Surface Soil for Radionuclides”

This standard describes sampling of surface soil for the purpose of obtaining a sample representative of a particular area for subsequent chemical analysis of selected radionuclides. The standard describes one acceptable approach to collect soil samples for radiochemical analysis. Guidance is provided for the collection of soil samples so as to be representative of the area. Soil provides a source material for the determination of selected radionuclides and serves as an integrator of the deposition of airborne materials. Soil sampling should not be used as the primary measurement system to demonstrate compliance with applicable radionuclides in air standards. Soil collected by

this practice and subsequent analysis is used to monitor radionuclide deposition of emissions from nuclear facilities.

- C999-90 (Reapproved 2000) “Standard Practice for Soil Sample Preparation for the Determination of Radionuclides”

This standard describes the preparation of surface soil samples collected for chemical analysis of radionuclides, particularly uranium and plutonium. The standard describes one acceptable approach to the preparation of soil samples for radiochemical analysis. Guidance is provided for the preparation of a homogeneous soil sample from ten composited core samples (aggregate weight of 4 to 5 kg) collected as to be representative of the area. Soil samples prepared for radionuclide analyzes by this practice are used to monitor fallout distribution from nuclear facilities. This practice is intended to produce a homogeneous sample from which a relatively small aliquot (10 g) may be drawn for radiochemical analyzes.

- C1000-00 “Standard Test Method for Radiochemical Determination of Uranium Isotopes in Soil by Alpha Spectrometry”

This test method covers the determination of uranium isotopes in soil.

The test method is designed to analyze 10 g of soil; however, the sample size may be varied to 50 g depending on the activity level. This test method may not be able to completely dissolve all forms of uranium in the soil matrix.

The lower limit of detection is dependent on count time, sample size, detector efficiency, background, and tracer yield. The chemical recovery averaged 78% in a single laboratory evaluation, and 66% in an interlaboratory collaborative study.

- C1001-00 “Standard Test Method for Radiochemical Determination of Plutonium in Soil by Alpha Spectroscopy”.

This test method covers the determination of plutonium in soils at levels of detection dependent on count time, sample size, detector efficiency, background, and tracer yield. This test method describes one acceptable approach to the determination of plutonium in soil.

This test method is designed for 10 g of soil, previously collected and treated as described in Practices C998 and C999, but sample sizes up to 50 g may be analyzed by this test method. This test method may not be able to completely dissolve all forms of plutonium in the soil matrix.

- C1109-98 “Standard Test Method for Analysis of Aqueous Leachates from Nuclear Waste Materials Using Inductively Coupled Plasma-Atomic Emission Spectrometry”

This test method is applicable to the determination of low concentration and trace elements in aqueous leachate solutions produced by the leaching of nuclear waste materials. The nuclear waste material may be a simulated (non-radioactive) solid waste form or an actual solid radioactive waste material. The leachate may be deionized water

or any natural or simulated leachate solution containing less than 1 % total dissolved solids. The analysis must be conducted with an inductively coupled plasma-atomic emission spectrometer. This test method may be used to determine concentrations of elements leached from nuclear waste materials (glasses, ceramics, cements) using an aqueous leachant. If the nuclear waste material is radioactive, a suitably contained and shielded ICP-AES spectrometer system with a filtered exit-gas system must be used, but no other changes in the test method are required.

- C1111-98 “Standard Test Method for Determining Elements in Waste Streams by Inductively Coupled Plasma-Atomic Emission Spectroscopy”

This test method covers the determination of trace, minor, and major elements in waste streams by inductively coupled plasma-atomic emission spectroscopy (ICP-AES) following an acid digestion of the sample. Waste streams from manufacturing processes of nuclear and nonnuclear materials can be analyzed. This test method is applicable to the determination of total metals. Results from this test method can be used to characterize waste received by treatment facilities and to formulate appropriate treatment recipes. The results are also usable in process control within waste treatment facilities. This test method is applicable only to waste streams that contain radioactivity levels which do not require special personnel or environmental protection. If the sample is a clear acidified solution, the elements are determined with no further pretreatment. If the sample contains undissolved solids, the elements are determined using an aliquot of the thoroughly mixed sample after a nitric acid digestion. This test method is useful for the determination of concentrations of metals in many waste streams from various nuclear and nonnuclear manufacturing processes. The test method is useful for characterizing liquid wastes and liquid wastes containing undissolved solids prior to treatment, storage, or stabilization. It has the capability for the simultaneous determination of up to 26 elements.

- C1205-97 (Reapproved 2002) “Standard Test Method for The Radiochemical Determination of Americium-241 in Soil by Alpha Spectrometry”

This method covers the determination of americium-241 in soil by means of chemical separations and alpha spectrometry. It is designed to analyze up to ten grams of soil or other sample matrices that contain up to 30 mg of combined rare earths. This method allows the determination of americium-241 concentrations from ambient levels to applicable standards. The values stated in SI units are to be regarded as standard.

- C1234-98 (Reapproved 2004) “Standard Practice for Preparation of Oils and Oily Waste Samples by High-Pressure, High-Temperature Digestion for Trace Element Determinations”

This test method describes a high-pressure, high-temperature digestion technique using the high-pressure asher (HPA) for preparation of oils and oily waste specimens for determination of up to 28 different elements by inductively coupled plasma-atomic emission plasma spectroscopy (ICP-AES), cold-vapor atomic absorption spectroscopy (CVAAS), and graphite furnace atomic absorption spectroscopy (GFAAS). Oily and high-percentage organic waste streams from nuclear and non-nuclear manufacturing processes can be successfully prepared for trace element determinations by ICP-AES,

CVAAS, and GFAAS. This method is applicable to the determination of total trace elements in these mixed wastes. Specimens prepared by this method can be used to characterize organic mixed waste streams received by hazardous waste treatment incinerators and for total element characterization of the waste streams.

This test method is applicable only to organic waste streams that contain radioactivity levels that do not require special personnel or environmental protection from radioactivity or other acute hazards.

A list of elements determined in oily waste streams is found in Table 1.

This test method has been used successfully to completely digest a large variety of oils and oily mixed waste streams from nuclear processing facilities. While the method has been used to report data on up to 28 trace elements, its success should not be expected for all analytes in every specimen. The overall nature of these oily wastes tends to be heterogeneous that can affect the results. Homogeneity of the prepared sample is critical to the precision and quality of the results. Some elements, notably silver and antimony, may be recovered on a semiquantitative basis, while most results are highly quantitative.

This procedure is designed to be applicable to samples whose preparation methods are not defined, or not suitable, by other regulatory procedures or requirements, such as the U.S. Environmental Protection Agency (EPA) SW-846 and EPA-600 4-79-020 publications. This digestion method is designed to provide a high level of accuracy and precision, but does not replace or override any regulatory requirements for sample preparation.

This method uses hazardous materials, operations, and equipment at high pressure (90-110 bars, 89-108 atm, or 1305-1595 lb/in.2) and high temperatures, up to 320°C, and therefore poses significant hazards if not operated properly. This standard does not purport to address all of the safety problems, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use. Specific hazard statements are given in Sections 10 and 11.

- C1255-93 (Reapproved 1999) “Standard Test Method for Analysis of Uranium and Thorium in Soils by Energy Dispersive X ray Fluorescence Spectroscopy”

This test method covers the energy dispersive X ray fluorescence (EDXRF) spectrochemical analysis of trace levels of uranium and thorium in soils. Any sample matrix that differs from the general ground soil composition used for calibration (that is, fertilizer or a sample of mostly rock) would have to be calibrated separately to determine the effect of the different matrix composition.

The analysis is performed after an initial drying and grinding of the sample, and the results are reported on a dry basis. The sample preparation technique used incorporates into the sample any rocks and organic material present in the soil. This test method of sample preparation differs from other techniques that involve tumbling and sieving the sample.

Linear calibration is performed over a concentration range from 20 to 1000 [ $\mu$ ]g per gram for uranium and thorium.

- C1284-00 “Standard Practice for Electrodeposition of the Actinides for Alpha Spectrometry”

This practice covers the preparation of separated actinide fractions for alpha spectrometry measurement. It is applicable to any of the actinides that can be dissolved in dilute ammonium sulfate solution. Examples of applicable actinide fractions would be the final elution from an ion exchange separation or the final strip from a solvent extraction separation.

- C1310-01 “Standard Test Method for Determining Radionuclides in Soils by Inductively Coupled Plasma-Mass Spectrometry Using Flow Injection Preconcentration”

This test method covers a procedure for measuring  $^{99}\text{Tc}$  and a procedure for measuring  $^{230}\text{Th}$  and  $^{234}\text{U}$  in soils. It is applicable to background soils and soils that have been contaminated by nuclear processes. It is intended as an alternative to radiochemical methods because it is faster, requires less labor, and produces less waste than many radiochemical methods. Samples are dried, ground, dissolved by fusion, and analyzed by inductively coupled plasma-mass spectrometry (ICP-MS). A sequential flow injection (FI) technique is used to provide lower detection limits than those obtained with direct aspiration into an ICP-MS, and, in the case of  $^{99}\text{Tc}$ , provides separation from interferences. The  $^{230}\text{Th}$  and  $^{234}\text{U}$  procedure also would work for  $^{232}\text{Th}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ , but the FI preconcentration usually is not required to measure these isotopes at the concentrations typically found in soils. The analysis system consists of a computer-controlled FI system attached to the nebulizer of an ICP-MS. The FI system concentrates the analytes by solid-phase extraction and, in the case of  $^{99}\text{Tc}$ , provides separation from interferences. The ICP-MS nebulizes the FI eluent into a radio frequency-supported argon plasma that produces, ideally, singly-charged atomic ions that are detected by mass spectrometry. Soil samples are dried, ground, and blended to achieve homogeneity. For  $^{99}\text{Tc}$  analysis, samples are fused with sodium peroxide and dissolved in nitric acid. For  $^{230}\text{Th}$  and  $^{234}\text{U}$  analysis, samples are fused with lithium metaborate and dissolved in nitric acid. The FI concentration step reduces detection limits by approximately a factor of ten compared to ICP-MS with conventional sample introduction.

- C1343-96 (Reapproved 2002) “Standard Test Method for Determination of Low Concentrations of Uranium in Oils and Organic Liquids by X ray Fluorescence”

This test method covers the steps necessary for the preparation and analysis by X ray fluorescence (XRF) of oils and organic solutions containing uranium.

The procedure is valid for those solutions containing 20 to 2000  $\mu\text{g}$  uranium/mL as presented to the spectrometer.

This test method requires the use of an appropriate internal standard. Care must be taken to ascertain that samples analyzed by this test method do not contain the internal standard or that this contamination, whenever present, has been corrected for mathematically. Such corrections are not addressed in this procedure. Care must be

taken that the internal standard and sample medium are compatible; that is, samples must be miscible with tri-*n*-butyl phosphate (TBP) and must not remove the internal standard from solution. Alternatively, a scatter line may be used as the internal standard.

— C1387-98 “Standard Guide for Determination of Technetium-99 in Soil”

This publication is intended to serve as a reference for laboratories wishing to perform Tc-99 analyzes in soil. Several options are given for selection of a tracer and for the method of extracting the Tc from the soil matrix. Separation of Tc from the sample matrix is performed using an extraction chromatography resin. Options are then given for the determination of the Tc-99 activity in the original sample. It is up to the user to determine which options are appropriate for use, and to generate acceptance data to support the chosen procedure.

Due to the various extraction methods available, various tracers used, variable detection methods used, and lack of certified reference materials for Tc-99 in soil, there is insufficient data to support a single method written as a standard method.

— C1402-98 “Standard Guide for High-Resolution Gamma ray Spectrometry of Soil Samples”

This standard covers the identification and quantitative determination of gamma ray emitting radionuclides in soil samples by means of gamma ray spectrometry. It is applicable to nuclides emitting gamma rays with energies greater than 20 keV. For typical gamma ray spectrometry systems and sample types, activity levels of about 5 Bq are measured easily for most nuclides, and activity levels as low as 0.1 Bq can be measured for many nuclides. It is not applicable to radionuclides that emit no gamma rays such as the pure beta-emitting radionuclides hydrogen-3, carbon-14, strontium-90, and Becquerel quantities of most transuranics. This guide does not address the in situ measurement techniques, where soil is analyzed in place without sampling. Use of a germanium semiconductor detector is necessary for high-resolution gamma ray measurements.

— C1416-99 “Standard Test Method for Uranium Analysis in Natural and Waste Water by X ray Fluorescence”

This test method applies for the determination of trace uranium content in natural and waste water. It covers concentration of U between 0.05 mg/L and 2mg/L.

— C1463-00 “Standard Practices for Dissolving Glass Containing Radioactive and Mixed Waste for Chemical and Radiochemical Analysis”

This standard includes three techniques suitable for dissolving glass samples that may contain nuclear wastes. These techniques used together or independently will produce solutions that can be analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES), inductively coupled plasma mass spectrometry (ICP-MS), atomic absorption spectrometry (AAS), radiochemical methods and wet chemical techniques for major components, minor components and radionuclides. One of the fusion practices and the microwave practice can be used in hot cells and shielded hoods after modification to meet local operational requirements. All three methods for

dissolving silicate matrix samples require the sample to be dried and ground to a fine powder. In the first method, a mixture of sodium tetraborate ( $\text{Na}_2\text{B}_4\text{O}_7$ ) and sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) is mixed with the sample and fused in a muffle for 25 min at  $950^\circ\text{C}$ . The sample is cooled, dissolved in hydrochloric acid, and diluted to appropriate volume for analyzes. The second method described in this standard involves fusion of the sample with potassium hydroxide (KOH) or sodium peroxide ( $\text{Na}_2\text{O}_2$ ) using an electric bunsen burner, dissolving the fused sample in water and dilute HCl, and making to volume for analysis. Dissolution of the sample using a microwave oven is described in the third method. The ground sample is digested in a microwave oven using a mixture of hydrofluoric (HF) and nitric ( $\text{HNO}_3$ ) acids. Boric acid is added to the resulting solution to complex excess fluoride ions. These three methods offer alternative dissolution methods for a total analysis of a glass sample for major, minor, and radionuclide components.

— C1475-00 “Standard Guide for Determination of Neptunium-237 in Soil”

This guide covers the determination of neptunium-237 in soil by means of radiochemical separations and alpha spectrometry. This guide provides options in the selection of sample preparation, separation, and measurement. Although neptunium-237 is not a component of global fallout, it is a product of production reactors and spent fuel processing. This guide is designed for analysis of ten grams of soil previously collected and treated in accordance with Practices C998 and C999. Larger-size samples of environmental soil may also be analyzed, as long as the concentrations of interferences such as uranium and thorium are at or near environmental concentrations. Depending on the choice of a sample dissolution method, all chemical forms of neptunium may not be completely solubilized. This guide should allow the determination of neptunium-237 concentrations from sub Becquerel per gram levels to applicable standards depending on count time, sample size, detector efficiency, background, and tracer yield.

— C1507-01 “Standard Test Method for Radiochemical Determination of Strontium-90 in Soil”

This test method is applicable to the determination of strontium-90 in soil at levels of detection dependent on count time, sample size, detector efficiency, background, and recovery.

This test method is designed for the analysis of ten grams of soil, previously collected and treated as described in Practices C 998 and C 999. This test method may not be able to completely dissolve all soil matrices

ASTM Standards under the Jurisdiction of C26.07 Waste Materials

— C1144-89 (Reapproved 1997) Standard Test Method for Splitting Tensile Strength for Brittle Nuclear Waste Forms

**Scope:** This test method is used to measure the static splitting tensile strength of cylindrical specimens of brittle nuclear waste forms. It provides splitting tensile-strength data that can be used to compare the strength of waste forms when tests are done on one size of specimen. The test method is applicable to glass, ceramic, and concrete waste

forms that are sufficiently homogeneous but not to coated-particle, metal-matrix, bituminous or plastic waste forms, or concretes with large scale heterogeneities.

**Summary:** A right-circular cylinder of the waste solid is loaded diametrically between two hardened, parallel bearing blocks positioned between the specimen and the two test machine platens, one of which is moving at a constant speed relative to the other. As the load increases, the resultant stress eventually reaches the fracture strength of the material, and the specimen splits along the vertical diameter, usually with some subsidiary fracture at other locations. The splitting tensile strength is calculated from the measured fracture load.

**Significance and Use:** The splitting tensile strength test can only be used on brittle waste materials such as ceramics, glass, concrete or other materials that also have tensile fracture strengths that are less than one third of the compression strengths. This test method is only applicable to brittle solids because these are the only materials that fail under a definable stress state for the test specimen geometry and loading. This test method does not determine the effects of time and environment on strength, nor does it address failure under long duration static loading. It can be used as a quality control check and for optimizing waste form processing.

— C1220-98 “Standard Test Method for Static Leaching of Monolithic Waste Forms for Disposal of Radioactive Waste”

This test method evaluates the relative chemical durability of simulated and radioactive monolithic waste forms such as glasses, ceramics, or cermets in various test solutions at temperatures  $<100^{\circ}\text{C}$  under low surface-area-to-volume (S/V) ratio conditions. It can be used to distinguish differences in the leaching behavior of various simulated or radioactive waste forms under the specific conditions of the test based on analysis of the test solution. Data from this test are used to calculate the normalized elemental mass loss from specimens exposed to aqueous solutions at temperatures  $<100^{\circ}\text{C}$ . Specimen surfaces may be altered during this test. These altered surfaces may be used to study the reaction of monolithic waste forms during static exposure to solutions. Specimens of known volume and geometric surface area are immersed in the reference leachants without agitation for defined time periods at defined temperatures. The surface-area-to-volume ratio (S/V) is held constant within 0.5 of  $10.0\text{ m}^{-1}$ . Three reference temperatures,  $40^{\circ}$ ,  $70^{\circ}$ , and  $90^{\circ}\text{C}$ , and a number of specific time periods are identified in a series of test matrices established to meet objectives that include evaluation of waste forms for comparative purposes. In the test method, three reference leachants are used: high-purity water and two solutions (silicate/bicarbonate and brine) that approximate fluids that the waste form may encounter in a geologic repository. In addition to the reference leachants, others may be used. The test is for application to simulated waste forms and to radioactive specimens. Data from this test may form part of the larger body of data that is necessary in the logical approach to long-term prediction of waste form behavior. In particular, solution concentrations and characterization and altered surfaces may be used in the testing of geochemical modeling codes. This test method excludes study of powdered or organic materials. This test method can be used as either a “characterization” or “accelerated” test under the protocol of ASTM International standard C 1174.

C1308-95 (Reapproved 2001) “Standard Test Method for Accelerated Leach Test for Diffusive Releases from Solidified Waste and a Computer Program to Model Diffusive, Fractional Leaching from Cylindrical Waste Forms”

This test method provides a method for accelerating the leach rate of solidified waste and determining if the release is diffusion-controlled. This test method is applicable to any material that does not degrade, deform, or change leaching mechanism during the test. If diffusion is the dominant leaching mechanism, then results of this test can be used to model long-term releases from waste forms. Diffusion can be confirmed as the leaching mechanism through the use of a computerized mathematical model for diffusion from the finite cylinder. The leaching mechanism should be verified as diffusion-controlled by a means other than analysis of the leach test data. For this purpose, analysis of post-leaching concentration profiles within the solid waste form is recommended. This test method is a semi-dynamic leach test, that is, the leachant is sampled and replaced periodically. Elevated temperatures, large volumes of leachant, frequent leachant changes, and small specimen size are used to obtain accelerated releases. This is a short-term test, requiring sampling on each of eleven or more successive days. Tests must be performed to demonstrate that the leaching mechanism does not change at the elevated test temperature.

- C1571-03 “Standard Guide for Characterization of Radioactive and/or Hazardous Wastes for Thermal Treatment”

This guide identifies methods to determine the physical and chemical characteristics of radioactive and/or hazardous wastes before a waste is processed at high temperatures, for example, vitrification into a homogeneous glass, glass-ceramic, or ceramic waste form. This includes waste forms produced by ex-situ vitrification (ESV), in-situ vitrification (ISV), slagging, plasma-arc, hot-isostatic pressing (HIP) and/or cold-pressing and sintering technologies. Note that this guide does not specifically address high temperature waste treatment by incineration but several of the analyzes described in this guide may be useful diagnostic methods to determine incinerator off-gas composition and concentrations.

The characterization of the waste(s) recommended in this guide can be used to (1) choose and develop the appropriate thermal treatment methodology, (2) determine if waste pretreatment is needed prior to thermal treatment, (3) aid in development of thermal treatment process control, (4) develop surrogate waste formulations, (5) perform treatability studies, (6) determine processing regions (envelopes) of acceptable waste form composition, (7) perform pilot scale testing with actual or surrogate waste, and/or (8) determine the composition and concentrations of off-gas species for regulatory compliance.

The analyzes discussed in this standard can be performed by a variety of techniques depending on equipment availability. For example, Gas Chromatograph Mass Spectrometry (GC/MS) can be used to measure the amount and type of off-gas species present. However, this standard assumes that such sophisticated equipment is unavailable for radioactive or hazardous waste service due to potential contamination of the equipment. The analyzes recommended are, therefore, the simplest and least costly analyzes that can be performed and still be considered adequate

This guide is applicable to radioactive and/or hazardous wastes including but not limited to, high-level wastes, low-level wastes, transuranic (TRU) wastes, hazardous wastes, mixed (hazardous and radioactive) wastes, heavy metal contaminated wastes, and naturally occurring or accelerator produced radioactive material (NARM or NORM) wastes. These wastes can be in the physical form of wet sludges, dried sludges, spent waste water filter aids, waste water filter cakes, incinerator ashes (wet or dry), incinerator blowdown (wet or dry), wastewaters, asbestos, resins, zeolites, soils, unset or unsatisfactory cementitious wastes forms in need of remediation, lead paint wastes, etc. and combinations of the above. This guide may not be applicable to piping, duct work, rubble, debris waste or wastes containing these components.

This guide references applicable test methods that can be used to characterize hazardous wastes, radioactive wastes, and heavy metal contaminated process wastes, waste forms, NARM or NORM wastes, and soils.

#### ASTM Standards under the Jurisdiction of C26.08 Quality Assurance, Statistical Applications, and Reference Materials

- C0970-87 “Standard Practice for Sampling Special Nuclear Materials in Multi-Container Lots”

This practice provides an aid in designing a sampling and analysis plan for the purpose of minimizing random error in the measurement of the amount of nuclear material in a lot consisting of several containers. The problem addressed is the selection of the number of containers to be sampled, the number of samples to be taken from each sampled container, and the number of aliquot analyzes to be performed on each sample.

This practice provides examples for application as well as the necessary development for understanding the statistics involved. The uniqueness of most situations does not allow presentation of step-by-step procedures for designing sampling plans. It is recommended that a statistician experienced in materials sampling be consulted when developing such plans.

- C1009-96 “Standard Guide for Establishing a Quality Assurance Program for Analytical Chemistry Laboratories Within the Nuclear Industry”

This guide covers the establishment of a quality assurance (QA) program for analytical chemistry laboratories within the nuclear industry. Reference to key elements of ANSI/ISO/ASQC Q9001-1994, Quality Systems, provides guidance to the functional aspects of analytical laboratory operation. When implemented, the recommended practices presented in this guide will provide a comprehensive QA program for the laboratory. The recommended practices are grouped by functions, which are the basic elements of a QA program.

- C1068-96 “Standard Guide for Qualification of Measurement Methods by a Laboratory within the Nuclear Industry”

This guide provides guidance for selecting, validating, and qualifying measurement methods when qualification is required for a specific program. The recommended practices presented in this guide provide a major part of a quality assurance program for

the laboratory data. Qualification helps to assure that the data produced will meet established requirements.

- C1128-01 “Standard Guide for Preparation of Working Reference Materials for Use in the Analysis of Nuclear Fuel Cycle Materials”

This guide covers the preparation and characterization of working reference materials (WRM) that are produced by a laboratory for its own use in the analysis of nuclear materials. Guidance is provided for establishing traceability of WRMs to certified reference materials by a defined characterization process. The guidance provided is generic; it is not specific for a given material.

This guide covers the preparation of WRMs from nuclear fuel cycle materials. These materials are compounds such as boron carbide, and cladding materials such as zirconium and stainless steel. The criteria governing the preparation of reliable WRMs are identified and discussed. Because this guide is generic, requirements and detailed information for specific nuclear materials are not given.

- C1156-95 “Standard Guide for Establishing Calibration for a Measurement Method Used to Analyze Nuclear Fuel Cycle Materials”

This guide provides the basis for establishing calibration for a measurement method typically used in an analytical chemistry laboratory analyzing nuclear materials. Guidance is included for such activities as preparing a calibration procedure, selecting a calibration standard, controlling calibrated equipment, and documenting calibration. The guide is generic and any required technical information specific for a given method must be obtained from other sources.

- C1188-91 “Standard Guide for Establishing a Quality Assurance Program for Uranium Conversion Facilities”

This guide provides guidance and recommended practices for establishing a comprehensive quality assurance program for uranium conversion facilities.

Quality assurance provides a planned and systematic approach for establishing practices to meet requirements of safe facility operation and product quality. In the operation of a uranium conversion facility there are many requirements established by regulatory bodies, codes, customers, and the facility itself. These requirements are identified by facility management and acted upon by various facility groups. Implementation of the practices described in this guide are intended to assist with compliance with these requirements. In the operation of a uranium conversion facility there is a potential for both chemical and radiological exposure to employees, the public, and the environment. This potential is reduced by implementation of the practices described in this guide. The development of this guide, as part of sound management practice, provides a means for ensuring consistency between facilities, and documentation and formalization of existing practices.

- C1210-96 “Standard Guide for Establishing a Measurement System Quality Control Program for Analytical Chemistry Laboratories within the Nuclear Industry”

This standard provides guidance for establishing and maintaining a measurement system quality control program. Guidance is provided for general program considerations, preparation of quality control samples, analysis of quality control samples, quality control data analysis, analyst qualification, measurement system calibration, measurement method qualification, and measurement system maintenance.

A laboratory quality assurance program is an essential program for laboratories within the nuclear industry. The basic elements of a laboratory quality assurance program are organization, quality assurance program, training and qualification, procedures, laboratory records, control of records, control of procurement, control of measuring equipment and materials, control of measurements, and deficiencies and corrective actions. This guide deals with the control of measurements aspect of the laboratory quality assurance program.

- C1215-92 “Standard Guide for Preparing and Interpreting Precision and Bias Statements in Test Method Standards Used in the Nuclear Industry”

This guide covers terminology useful for the preparation and interpretation of precision and bias statements. In formulating precision and bias statements, it is important to understand the statistical concepts involved and to identify the major sources of variation that affect results. This guide provides a brief summary of these concepts. To illustrate these statistical concepts and to demonstrate some sources of variation, a hypothetical data set is analyzed.

This guide is intended as an aid to understanding the statistical concepts used in precision and bias statements. There is no intention that this be a self-contained introduction to statistics. Since many analysts have no formal statistical training, it is advised that a trained statistician be consulted for further clarification if necessary.

- C1297-95 “Standard Guide for Qualification of Laboratory Analysts for the Analysis of Nuclear Fuel Cycle Materials”

This guide covers the qualification of analysts to perform chemical analysis or physical measurements of nuclear fuel cycle materials. The guidance is general in that it is applicable to all analytical methods, but must be applied method by method. Also, the guidance is general in that it may be applied to initial qualification or requalification

This guide describes an approach to analyst qualification that is designed to be used in conjunction with a rigorous program for the qualification and control of the analytical measurement system. This requires an existing data base which defines the characteristics (precision and bias) of the system in routine use. The process described here is intended only to qualify analysts when such a data base exists and the method is in control. The qualification activities described in this guide assume that the analyst is already proficient in general laboratory operations. The training or other activities that developed this proficiency are not covered in this guide.

### ASTM Standards under the Jurisdiction of D34.01 on Physical and Chemical Characterization

- D5792-02 “Standard Practice for Generation of Environmental Data Related to Waste Management Activities: Development of Data Quality Objectives”
- D6699-01 “Standard Practice for Sampling Liquids Using Bailers”
- D6759 “Standard Practice for Sampling Liquids Using Grab and Discrete Depth Samplers”

### ASTM Standards under the Jurisdiction of C26.09 Nuclear Processing

- C1062-00 “Standard Guide for Design, Fabrication, and Installation of Nuclear Fuel Dissolution Facilities”

This guide establishes criteria and procedures for the design, fabrication, and installation of nuclear fuel dissolution facilities. The guide applies to all processing steps or operations beyond fuel shearing operations (which are not covered) up to and including the accountability vessel for the dissolver solution. This guide does not address special dissolution processes that may require substantially different equipment or pose different hazards than those associated with the fuel types currently used in Pressurized Water Reactors (PWR), Boiling Water Reactors (BWR), Pressurized Heavy Water Reactors (PHWR) and Heavy Water Reactors (HWR). The guide does not address the design and fabrication of continuous dissolvers. The purpose of this guide is to provide information that will help to ensure that nuclear fuel dissolution facilities are conceived, designed, fabricated, constructed, and installed in an economic and efficient manner. This guide will help facilities meet the intended performance functions, eliminate or minimize the possibility of nuclear criticality and provide for the protection of both the operator personnel and the public at large under normal and abnormal (emergency) operating conditions as well as under credible failure or accident conditions.

- C1217-00 “Standard Guide for Design of Equipment for Processing Nuclear and Radioactive Materials”

This guide covers equipment used in shielded cell or canyon facilities for the processing of nuclear and radioactive materials. The guide provides conditions and practices that have been found necessary to ensure against or to minimize the failures and outages of equipment used for radioactive service. It is intended that equipment designed, fabricated, procured, or obtained by transfer or adaptation and re-use of existing equipment, and installed in accord with the standard meet or exceed statutory, regulatory, and safety requirements for that equipment under the applicable operating and service conditions.

### ASTM Standards under the Jurisdiction of C26.10 Non Destructive Assay Methods

- C1030-95 (Reapproved 2001) “Standard Test Method for Determination of Plutonium Isotopic Composition by Gamma ray Spectrometry”

**Scope:** This test method is applicable to the determination of isotopic abundances in isotopically homogeneous Pu-bearing materials. The procedure is applicable to sample

sizes ranging from a few tenths of a gram up to the maximum sample weight allowed by criticality limits. This test method has been demonstrated in routine use for isotopic abundances ranging from 94 to 70 %  $^{239}\text{Pu}$ . This test method has also been employed for isotopic abundances outside this range.

**Summary:** Relative intensities of gamma rays from a plutonium sample are determined from a gamma ray spectrum obtained with a high-resolution Ge detector. The relative detection efficiency,  $e$ , is a function of gamma ray energy and results from the combined effects of detector response, attenuation due to absorbers and container walls, and self-absorption within the sample for gamma rays of differing energies. The relative detection efficiencies are determined for each sample from the observed gamma spectrum.

**Significance and use:** The determination of isotopic composition by gamma ray spectrometry is a nondestructive technique and when used with other nondestructive techniques, such as calorimetry or neutron coincidence counting, can provide a totally destructive plutonium assay necessary for material accountancy and safeguards needs. Since gamma ray spectrometry systems are typically automated, the routine use of the test method is fast, reliable, and is not labor intensive. Since the test method is nondestructive, requiring no sample preparation, it does not create waste disposal problems. This test method assumes that the isotopic composition of plutonium in the sample being measured is homogeneous. The isotopic composition of a given batch or sample of plutonium is an attribute of that sample and, once determined, can be used in subsequent inventory measurements to verify the identity of a sample within the measurement uncertainties.

- C1133-96 (Reapproved 2003) “Standard Test Method for Nondestructive Assay of Special Nuclear Material in Low Density Scrap and Waste by Segmented Passive Gamma ray Scanning”

**Scope:** This test method covers the nondestructive assay of gamma ray emitting special nuclear materials (SNMs), most commonly  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Am}$ , in low-density scrap or waste, packaged in cylindrical containers. High-resolution gamma ray spectroscopy is used to detect and measure the nuclides of interest and to measure and correct for gamma ray attenuation in vertical segments of the container. Corrections are also made for counting losses occasioned by signal processing limitations. Two conditions must be met to optimize SGS assay results as follows: The particles containing the nuclides of interest must be small to minimize self absorption of emitted gamma radiation. The mixture of material within each item segment must be sufficiently uniform to apply an attenuation correction factor, computed from a measurement of gamma ray transmission through the segment. The assay technique may be applicable to loadings of from one to several hundred grams of nuclide, with more restricted ranges to be applicable depending on specific packaging and counting equipment considerations. Measured transmission values must be available to permit valid attenuation corrections.

**Summary:** The assay of the nuclides of interest is accomplished by measuring the intensity of a characteristic gamma ray from each nuclide. Corrections are made for count rate-related losses and attenuation by the item. Comparison to similarly corrected gamma ray intensities, observed during the measurement of appropriate calibration

materials, provides the relationship between observed gamma ray intensity and nuclide content. The assay item is rotated about its vertical axis and scanned segment by segment along that axis, thereby reducing the effects of nonuniformity in both matrix density and nuclide distribution. The average linear attenuation coefficient of each vertical segment is calculated by measurement of the transmitted intensity of an external gamma ray source. The corrected gamma ray count rates for the nuclides of interest are determined on a segment-by-segment basis. At the completion of the measurement of all segments, count rates are summed, and mass values for the nuclides of interest in the entire container are calculated based on comparisons to appropriate calibration materials.

**Significance and use:** Segmented gamma ray scanning provides a nondestructive means of measuring the nuclide content of scrap and waste where the specific nature of the matrix and the chemical form and relationship between the nuclide and matrix may be unknown. The procedure can serve as a diagnostic tool that provides a vertical profile of transmission and nuclide concentration within the item. The procedure is highly automated and requires little operator interaction. Sample preparation is generally limited to good waste/scrap segregation practices that produce relatively homogeneous items that are required for any successful waste/inventory management and assay scheme, regardless of the measurement method used.

- C1207-97 (Reapproved 2003) “Standard Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting”

**Scope:** This test method describes the nondestructive assay of scrap or waste for plutonium content using passive thermal neutron coincidence counting. This test method provides rapid results and can be applied to a variety of carefully sorted materials in containers as large as 208-L drums. The test method applies to measurements of  $^{238}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{242}\text{Pu}$  and has been used to assay items whose total plutonium content ranges from 0.01 to 6000 g. This test method requires knowledge of the relative abundances of the plutonium isotopes to determine the total plutonium mass. This test method may not be applicable to the assay of scrap or waste containing other spontaneously fissioning nuclides and may give biased results for measurements of containers that include large amounts of hydrogenous materials.

**Summary:** The even mass isotopes of plutonium fission spontaneously. On the average, two or more neutrons are emitted per fission event. The number of these coincident neutrons detected by the instrument is correlated to the quantity of even mass isotopes of plutonium in the assay item,  $m_{\text{eff}}$ . The total plutonium mass is determined from the known plutonium isotopic ratios and the measured quantity of even mass isotopes. The shift register technology is intended to correct for the effects of accidental neutrons. Other factors which may affect the assay are multiplication and matrix components with large ( $\alpha, n$ ) reaction rates, neutron absorbers, or moderators. Corrections for these effects are often not possible from the measurement data alone, consequently assay items are sorted into material categories or additional information is used to obtain the best assay result.

**Significance and use:** This test method is useful for determining the plutonium content of scrap and waste in containers as large as 208-L (55-gal) drums. Total plutonium

content ranges from 10 mg to 6 kg . The upper limit may be restricted to smaller mass values depending on specific matrix, calibration material, criticality safety, or counting equipment considerations. This test method is applicable for shipper/receiver confirmatory measurements, nuclear material diversion detection, and International Atomic Energy Agency attributes measurements. This test method should be used in conjunction with a scrap and waste management plan that segregates scrap and waste assay items into material categories according to some or all of the following criteria: bulk density, the chemical forms of the plutonium and the matrix, americium to plutonium isotopic ratio, and hydrogen content. Packaging for each category should be uniform with respect to size, shape, and composition of the container. Each material category will require calibration standards and may have different plutonium mass limits. For both benign matrix and matrix specific measurements, the method assumes the calibration reference materials match the items to be measured with respect to parameters that affect the measurement. This method is recommended for measurements on items with homogeneous contents.

- C1221-92 (Reapproved 1998) “Standard Test Method for Nondestructive Analysis of Special Nuclear Materials in Homogeneous Solutions by Gamma ray Spectrometry”

**Scope:** This test method is applicable to the determination of the concentration of gamma ray emitting special nuclear materials dissolved in homogeneous solutions. The test method corrects for gamma ray attenuation by the sample and its container by measurement of the transmission of a beam of gamma rays from an external source. Two sample geometries, slab and cylinder, are considered. The sample container that determines the geometry may be either a removable or a fixed geometry container. This test method is limited to sample containers having walls or a top and bottom of equal transmission through which the gamma rays from the external transmission correction source must pass. This test method is typically applied to radionuclide concentrations ranging from a few mg/L to several hundred g/L. The assay range will be a function of the specific activity of the nuclide of interest, the physical characteristics of the sample container, counting equipment considerations, assay gamma ray energies, sample matrix, gamma ray branching ratios, and interferences.

**Summary:** The gamma ray spectrum of a portion of solution is obtained with a collimated, high resolution gamma ray detector. A correction factor for gamma ray attenuation in the sample and its container is determined from the measurement of the transmitted intensity of an external gamma ray source. The gamma rays from the external source have energies close to those of the assay gamma rays emitted from the sample. The relationship between the measured gamma ray intensity and the nuclide concentration (the calibration constant) is determined by use of appropriate standards.

**Significance and use:** This test method is a nondestructive means of determining the nuclide concentration of a solution for special nuclear material accountancy, nuclear safety, and process control. It is assumed that the nuclide to be analyzed is in a homogeneous solution. The transmission correction makes the test method independent of matrix and useful over several orders of magnitude of nuclide concentrations. However, a typical configuration will normally span only two to three orders of magnitude. The test method assumes that the sample-detector geometry is the same for all measured items. This can be accomplished by requiring that the liquid height in the

sidelooking geometry exceed the detector field of view defined by the collimator. For the uplooking geometry, a fixed sample fill height must be maintained and vials of identical radii must be used unless the vial radius exceeds the field of view defined by the collimator. Since gamma ray systems can be automated, the test method can be rapid, reliable and not labor intensive. This test method may be applicable to in-line or off-line situations.

- C1268-94 (Reapproved 2000) “Standard Test Method for Quantitative Determination of Americium 241 in Plutonium by Gamma ray Spectrometry”

**Scope:** This test method covers the quantitative determination of americium 241 by gamma ray spectrometry in plutonium nitrate solution samples that do not contain significant amounts of radioactive fission products or other high specific activity gamma ray emitters. This test method can be used to determine the americium 241 in samples of plutonium metal, oxide and other solid forms, when the solid is appropriately sampled and dissolved.

**Summary:** An aliquot of the sample that contains about 10 to 100 ng of americium 241 is analyzed by measuring the intensity of the characteristic 59.5 keV gamma ray emitted by americium 241. Multiple sample geometries may be used if an appropriate calibration for each geometry is made. The sample geometry must be reproducible. This includes the physical characteristics of the sample container, the positioning of the sample, and the volume of sample viewed by the gamma ray detector. The necessity of dead time and pulse pile-up corrections can be reduced by sample dilution to control count rates. A correction is made for the contribution to the 59.5 keV intensity due to gamma rays produced in the decay of uranium 237. The relationship between the measured gamma ray intensity and the americium 241 content is determined by the use of appropriate standards.

**Significance and use:** This test method allows the determination of americium 241 in a plutonium solution without separation of the americium from the plutonium. It is generally applicable to any solution containing americium 241. The americium 241 in solid plutonium materials may be determined when these materials are dissolved. When the plutonium solution contains unacceptable levels of fission products or other materials, this method may be used following a tri-n-octylphosphine oxide (TOPO) extraction, ion exchange or other similar separation techniques. This test method is less subject to interferences from plutonium than alpha counting since the energy of the gamma ray used for the analysis is better resolved from other gamma rays than the alpha particle energies used for alpha counting. The minimal sample preparation reduces the amount of sample handling and exposure to the analyst. This test method is applicable only to homogeneous solutions. This test method is not suitable for solutions containing solids. Solutions containing as little as  $1 \times 10^{-5}$  g/L americium 241 may be analyzed using this method. The lower limit depends on the detector used and the counting geometry. Solutions containing high concentrations may be analyzed following an appropriate dilution.

- C1316-01 “Standard Test Method for Nondestructive Assay of Nuclear Material in Scrap and Waste by Passive-Active Neutron Counting Using a  $^{252}\text{Cf}$  Shuffler”

**Scope:** This test method covers the nondestructive assay of scrap and waste for uranium and plutonium content using a  $^{252}\text{Cf}$  shuffler. Shuffler measurements provide rapid results and can be applied to a variety of matrix materials in containers as large as 208-litre drums. Corrections are made for the effects of matrix material. This test method has been used to assay items containing uranium, plutonium, or both. Applications of this test method include measurements for safeguards, accountability, TRU, and U waste segregation, disposal, and process control purposes. This test method uses passive neutron coincidence counting to measure  $^{238}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{242}\text{Pu}$ . It has been used to assay items with plutonium contents between 0.03 g and 1000 g. It could be used to measure other spontaneously fissioning isotopes. This test method uses neutron irradiation with a moveable californium source and counting of the delayed neutrons from the induced fissions to measure  $^{235}\text{U}$ . It has been used to assay items with  $^{235}\text{U}$  contents between 0.1 g and 1000 g. It could be used to assay other fissionable isotopes.

**Summary:** This test method consists of two distinct modes of operation: passive and active. The instrument that performs the active mode measurement is referred to as a “shuffler” due to the motion of the  $^{252}\text{Cf}$  source. This test method usually relies on passive neutron coincidence counting to determine the plutonium content of the item, and active neutron irradiation followed by delayed neutron counting to determine the uranium content. Either corrections are made for the effects of neutron absorbers and moderators in the matrix, or a matrix-specific calibration is used. The effect that needs correction is the increase or decrease in the neutron signal caused by the matrix. Corrections are made for electronic deadtime, neutron background, and the  $^{252}\text{Cf}$  source decay. The active mode also induces fissions in plutonium if it is present in the assay item. The passive measurement of plutonium can be used to correct the active measurement of  $^{235}\text{U}$  for the presence of plutonium. Calibrations are based on measurements of well-documented reference materials.

**Significance and use:** This test method is used to determine the uranium and plutonium content of scrap and waste in containers. Measurement count times have been 100 to 1000 s. The following limits may be further restricted depending upon specific matrix, calibration material, criticality safety, or counting equipment considerations. The passive measurement has been applied to benign matrices in 208-litre drums with plutonium content ranging from 30 mg to 1 kg. The active measurement has been applied to benign matrices in 208-litre drums with  $^{235}\text{U}$  content ranging from 100 mg to 1 kg. This test method can be used to demonstrate compliance with the radioactivity levels specified in safeguards, waste, disposal, and environmental regulations. This test method can detect diversion attempts that use bulk neutron shielding to encapsulate nuclear material.

- C1455-00 “Standard Guide for Nondestructive Assay of Special Nuclear Material Holdup Using Gamma ray Spectroscopic Methods”

**Scope:** This guide addresses methods used to prepare for and to perform, using gamma ray measurements, the nondestructive assay (NDA) of radioisotopes, for example,  $^{235}\text{U}$ , or  $^{239}\text{Pu}$ , remaining as holdup in nuclear facilities. Holdup occurs in facilities where nuclear material is processed. This guide includes the measurement of holdup of Special Nuclear Material (SNM) in places where holdup may occur, such as in process equipment, and in exhaust ventilation systems. This guide includes information useful

for management planning, selection of equipment, consideration of interferences, measurement program definition, and the utilization of resources. Nuclear material held up in pipes, ductwork, gloveboxes, heavy equipment, and so forth, usually is distributed in a diffuse and irregular manner. It is difficult to define the measurement geometry, identify the form of the material, and measure it without interference from adjacent sources of radiation. A scientific knowledge of radiation sources and detectors, calibration procedures, geometry and error analysis also is needed.

**Summary:** Holdup measurements range from the assay of a single item to routine measurement of a piece of equipment, to an extensive campaign of determining the total SNM in-process inventory for a processing plant. Holdup measurements differ from other nondestructive measurement methods in that the assays are performed in situ on equipment associated with the process. Often, the chemical form and geometric distribution of the SNM are not known. These unique challenges require for each measurement a specific definition of what is expected from the assay, specific information about the item or items to be assayed, design of the assay, and special preparation for the assay. The amount of effort expended and level of detail attained for each of these preparatory activities is dependent on both assay requirements and available resources. Information must be gathered concerning the item or items to be assayed and the level of effort needed to meet the holdup measurement requirements. Factors to be considered include the geometric configuration of the item or process equipment to be assayed, location of the equipment in the facility, attenuating materials, sources of background or interferences, safety considerations (both radiological and industrial) associated with the assay, plus the personnel and equipment needed to complete the assay.

**Significance and use:** This guide can apply to the measurement of holdup in equipment whose gamma ray absorption properties may be measured or estimated. These methods may be adequate to accurately measure items with complex distributions of holdup and attenuating material, however, the results are subject to larger measurement uncertainties than measurements of less complex distributions of holdup.

- C1458-00 “Standard Test Method for Nondestructive Assay of Plutonium, Tritium and <sup>241</sup>Am by Calorimetric Assay”

**Scope:** This test method describes the nondestructive assay (NDA) of plutonium, tritium, and <sup>241</sup>Am using heat flow calorimetry. For plutonium the range of applicability corresponds to < 1 g to > 2000 g quantities while for tritium the range extends from 0.001 g to > 10 g. This test method can be applied to materials in a wide range of container sizes up to 50L. It has been used routinely to assay items whose thermal power ranges from 0.001 W to 135 W. This test method requires knowledge of the relative abundances of the plutonium isotopes and the <sup>241</sup>Am/Pu mass ratio to determine the total plutonium mass. This test method provides a direct measure of tritium content. This test method provides a measure of <sup>241</sup>Am either as a single isotope or mixed with plutonium.

**Summary:** The item is placed in the calorimeter measurement chamber and the total heat flow at equilibrium, that is, the thermal power, from the item is determined by temperature sensors and associated electronic equipment. The thermal power emitted by

a test item is directly related to the quantity of radioactive material in it. The total power generated by ionizing radiation absorbed in the item is captured by the calorimeter. The mass of plutonium, tritium, or  $^{241}\text{Am}$  ( $m$ ) is calculated from the measured thermal power of an item ( $W_i$ ) divided by the effective specific power for that item. The effective specific power is calculated from the isotopic composition of the item.

**Significance and use:** This test method is presently the most accurate NDA technique for the assay of many physical forms of plutonium. Isotopic measurements by gamma ray spectroscopy or destructive analysis techniques are part of the test method when it is applied to the assay of plutonium. Calorimetry has been applied to a wide variety of Pu-bearing solids including metals, alloys, oxides, fluorides, mixed Pu-U oxides, mixed oxide fuel pins, waste, and scrap, for example, ash, ash heels, salts, crucibles, and graphite scarfings) . The test method is the most accurate NDA method for the measurement of tritium. For many physical forms of tritium compounds calorimetry is the only practical measurement technique available. Unlike other NDA techniques no physical standards representative of the materials being assayed are required for the test method. The test method is largely independent of the elemental distribution of the nuclear materials in the matrix.

- C1490-01 “Standard Guide for the Selection, Training and Qualification of Nondestructive Assay (NDA) Personnel”

**Scope:** This guide contains good practices for the selection, training, qualification, and professional development of personnel performing analysis, calibration, physical measurements, or data review using nondestructive assay equipment, methods, results, or techniques. The guide also covers NDA personnel involved with NDA equipment setup, selection, diagnosis, troubleshooting, or repair. Selection, training, and qualification programs based on this guide are intended to provide assurance that NDA personnel are qualified to perform their jobs competently. This guide presents a series of options but does not recommend a specific course of action.

**Significance and use:** The process of selection, training and qualification of personnel involved with NDA measurements is one of the quality assurance elements for an overall quality NDA measurement program. This guide describes an approach to selection, qualification, and training of personnel that is to be used in conjunction with other NDA QA program elements. The selection, qualification and training processes can vary and this guide provides one such approach. The qualification activities described in this guide assume that NDA personnel are already proficient in general facility operations and safety procedures. The training and activities that developed this proficiency are not covered in this guide. This guide describes a basic approach and principles for the qualification of NDA professionals and technical specialists and operators. A different approach may be adopted by the management organization based on its particular organization and facility specifics. However, if a variation of the approach of this guide is applied, the resulting selection, training, and qualification programs must meet the requirements of the facility quality assurance program and should provide all the applicable functions contained in this guide. This guide may be used as an aid in the preparation of a Training Implementation Plan (TIP) for the Transuranic Waste Characterization Program (TWCP). Requirements for TWCP

training include but are not limited to: 10 CFR 830.120, DOE Order 5480.20A, DOE QAPD CAO-94-1012, TWCP-QAPP CAO-94-1010, TWCP-QAPjP and the TUM.

- C1493-01 “Standard Test Method for Non-Destructive Assay of Nuclear Material in Waste by Passive and Active Neutron Counting Using a Differential Die-Away System”

**Scope:** This test method covers a system that performs nondestructive assay (NDA) of uranium or plutonium, or both, using the active, differential die-away technique (DDT), and passive neutron coincidence counting. Results from the active and passive measurements are combined to determine the total amount of fissile and spontaneously-fissioning material in drums of scrap or waste as large as 208 L. Corrections are made to the measurements for the effects of neutron moderation and absorption, assuming that the effects are averaged over the volume of the drum and that no significant lumps of nuclear material are present. These systems are most widely used to assay low-level and transuranic waste, but may also be used for the measurement of scrap materials. In the active mode, the system measures fissile isotopes such as  $^{235}\text{U}$  and  $^{239}\text{Pu}$ . The neutrons from a pulsed, 14-MeV neutron generator are thermalized to induce fission in the assay item. Between generator pulses, the system detects prompt-fission neutrons emitted from the fissile material. This method is called the differential die-away technique. In the passive mode, the system detects time coincident neutrons emitted from spontaneously fissioning isotopes. The primary isotopes measured are  $^{238}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{242}\text{Pu}$ ; however, the system may be adapted for use on other spontaneously-fissioning isotopes as well. This test method will give biased results when the waste form does not meet the calibration specifications and the measurement assumptions presented in this test method regarding the requirements for a homogeneous matrix, uniform source distribution, and the absence of nuclear material lumps, to the extent that they effect the measurement.

**Summary:** This test method addresses a system that performs active differential die-away and passive neutron coincidence counting. Typically, the active mode is performed prior to the passive mode. A 208 L drum is placed inside the chamber and rotated continuously during the measurement. The active mode is performed by interrogating the drum with neutrons from a pulsed neutron generator for 40 to 200 s. The passive mode is performed using a counting interval of 200 to 1000 s. If the isotopic ratios as well as the relative responses are known for individual radionuclides, the active and passive modes can be used to give independent measurements of the total plutonium mass. The system can also be operated only in the passive mode to measure the plutonium content of scrap or waste, or only in the active mode for measurement of uranium.

**Significance and use:** This test method is useful for quantifying fissile (for example,  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$ ) and spontaneously-fissioning nuclei (for example,  $^{238}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{242}\text{Pu}$ ,  $^{244}\text{Cm}$ ,  $^{248}\text{Cm}$ , and  $^{252}\text{Cf}$ ) in waste and scrap drums. Total elemental mass of the radioactive materials can be calculated if the relative abundances of each radionuclide are known. Typically, this test method is used to measure one fissile isotope (for example,  $^{235}\text{U}$  or  $^{239}\text{Pu}$ ). This test method can be used to segregate low level and transuranic waste at the 100 nCi/g concentration level currently required to meet the DOE Waste Isolation Pilot Plant (WIPP) waste acceptance criterion. For uranium-bearing items, the DDT can measure the  $^{235}\text{U}$  content in the range from 0.02 to over 100

g. Normally the assay of items bearing only uranium is performed using matrix-specific calibrations to account for the effect of the matrix on the active signal. For plutonium-bearing items, the DDT method measures the  $^{239}\text{Pu}$  content in the range between 0.01 and 20 g. The passive mode is capable of assaying spontaneously-fissioning nuclei, over a nominal range from 0.05 to 15 g of  $^{240}\text{Pu}$ , or equivalent. The passive mode can also be used to measure large (for example, kg) quantities of  $^{238}\text{U}$ .

— C1500-02 “Standard Test Method for Nondestructive Assay of Plutonium by Passive Neutron Multiplicity Counting”

**Scope:** This test method describes the nondestructive assay of plutonium in forms such as metal, oxide, scrap, residue, or waste using passive neutron multiplicity counting. This test method provides rapid results that are usually more accurate than conventional neutron coincidence counting. The method can be applied to a large variety of plutonium items in various geometries in cans, 208-L drums, or 1900-L Standard Waste Boxes. It has been used to assay items whose plutonium content ranges from 1 g to 1000's of g. This test method is primarily intended to address the assay of  $^{240}\text{Pu}$ -effective by moments-based multiplicity analysis using shift register electronics and high efficiency neutron counters specifically designed for multiplicity analysis. This test method requires knowledge of the relative abundances of the plutonium isotopes to determine the total plutonium mass.

**Summary:** The item is placed in the sample chamber or “well” of the multiplicity counter, and the emitted neutrons are detected by the  $^3\text{He}$  tubes that surround the well. The detected neutron multiplicity distribution is processed by the shift register electronics package to obtain the number of neutrons of each multiplicity in the (R + A) and (A) gates. The first three moments of the (R + A) and (A) multiplicity distributions are computed to obtain the singles (or totals), the doubles (or reals), and the triples. Using these three calculated values, it is possible to solve for 3 unknown item properties, the  $^{240}\text{Pu}$ -effective mass, the self-multiplication, and the  $\alpha$  ratio. The total plutonium mass is then determined from the known plutonium isotopic ratios and the  $^{240}\text{Pu}$ -effective mass. Corrections are routinely made for neutron background, cosmic ray effects, small changes in detector efficiency with time, and electronic deadtimes. Optional algorithms are available to correct for the biases caused by spatial variations in self-multiplication or changes in the neutron die-away time.

**Significance and use:** This test method is useful for determining the plutonium content of items such as impure Pu oxide, mixed Pu/U oxide, oxidized Pu metal, Pu scrap and waste, Pu process residues, and weapons components. Measurements made with this test method may be suitable for safeguards or waste characterization requirements. A significant feature of neutron multiplicity counting is its ability to capture more information than neutron coincidence counting because of the availability of a third measured parameter, leading to reduced measurement bias for most material categories. This feature also makes it possible to assay some in-plant materials that are not amenable to conventional coincidence counting, including moist or impure plutonium oxide, oxidized metal, and some categories of scrap, waste, and residues. Calibration for many material types does not require representative standards. Thus, the technique can be used for inventory verification without calibration standards, although measurement bias may be lower if representative standards were available. Bias in passive neutron

multiplicity measurements is related to deviation from the “point model” such as variations in detection efficiency, matrix composition, or distribution of nuclear material in the item’s interior.

— C1514-02 “Standard Test Method for Measurement of  $^{235}\text{U}$  Fraction using the Enrichment Meter Principle”

**Scope:** This test method covers the quantitative determination of the fraction of  $^{235}\text{U}$  in uranium using measurement of the 185.7 keV gamma ray produced during the decay of  $^{235}\text{U}$ . This test method is applicable to items containing homogeneous uranium-bearing materials of known chemical composition in which the compound is considered infinitely thick with respect to 185.7 keV gamma rays. This test method can be used for the entire range of  $^{235}\text{U}$  fraction, from depleted (0.2 %  $^{235}\text{U}$ ) to very highly enriched (97.5 %  $^{235}\text{U}$ ).

**Summary:** The test method consists of measuring the emission rate of 185.7 keV gamma rays from an item and correlating that emission rate with the enrichment of the uranium contained in the item. Calibration is achieved using reference materials of known enrichment. Corrections are made for attenuating materials present between the uranium-bearing material and the detector and for chemical compounds different from the calibration reference materials used for calibration. The measured items must completely fill the field of view of the detector, and must contain a uranium-bearing material that is infinitely thick with respect to the 185.7 keV gamma ray.

**Significance and use:** The enrichment meter principle provides a nondestructive measurement of the  $^{235}\text{U}$  fraction of uranium-bearing items. Sampling is not required and no waste is generated, minimizing exposure to hazardous materials and resulting in reduced sampling error. Use of a low resolution detector (e.g., NaI detector) to measure uranium with  $^{235}\text{U}$  fraction approximately 10 % which is contained in a thin-walled container can provide a rapid (typically 100 s), easily portable measurement system with precision of 0.6 % and bias of less than 1 %. Use of a high resolution detector (e.g., high-purity germanium) can provide measurement with a precision better than 0.2 % and a bias less than 1 % within a 300-sec measurement time when measuring uranium with  $^{235}\text{U}$  fraction in the range of 0.711 % to 4.46 % which is contained in thin-walled containers. In order to obtain optimum results using this method, the chemical composition of the item must be well known, the container wall must permit transmission of the 185.7 keV gamma ray, and the uranium-bearing material within the item must be infinitely thick with respect to the 185.7 keV gamma ray. Items must be homogeneous with respect to both  $^{235}\text{U}$  fraction and chemical composition. The uranium-bearing materials in the measured items and calibration reference materials used for calibration must fill the detector field of view.

Standards currently under development by C26.10

— C1592-04 Standard Guide for Nondestructive Assay Measurements

**Scope:** This guide is a compendium of Good Practices for performing measurements of radioactive material using nondestructive assay instruments. The primary purpose of the guide is to assist its users in arriving at quality NDA results, i.e., results that satisfy the

end user's needs. This is accomplished by providing an acceptable and uniform basis for the collection, analysis, comparison, and application of data. The recommendations are not compulsory or pre requisites to achieving quality NDA measurements, but are considered contributory in most areas. This guide applies to the use of NDA instrumentation for the measurement of nuclear materials by the observation of spontaneous or stimulated nuclear radiations, including photons, neutrons, or the flow of heat. Recommended calibration, operating, and assurance methods represent guiding principles based on current NDA technology. This guide is to be used as a reference, and to supplement the critical thinking, professional skill, expert judgment, and experimental test and verification needed to ensure that the instrumentation and methods have been properly implemented. When applied to measurement of waste, this guide should be used in conjunction with a waste management plan that segregates the contents of assay items into material categories according to some or all of the following criteria: bulk density of the waste, chemical forms of the plutonium or uranium and matrix, (alpha, n) neutron intensity, hydrogen (moderator) and absorber content, thickness of fissile mass(es), and the assay item container size and composition. Each matrix may require a different set of calibration standards and may have different mass calibration limits. The effect on the quality of the assay (i.e., minimizing precision and bias) can significantly depend on the degree of adherence to this waste management plan.

**Summary:** Good NDA measurement practices are described in this guide. Nondestructive assay measurements are typically performed when the items measured or goals of the measurement program favor NDA over destructive radio assay analysis. NDA is typically favored when collecting a representative sample of the item is difficult or impractical (e.g., scrap and waste items), personnel exposure would be significant, spread of contamination from sampling would occur, generation of secondary waste is undesirable, the weight and/or tare weight of the item cannot easily be determined (e.g., in place process equipment), rapid turn around of the measurement results is needed, or the NDA measurement is significantly less expensive than the equivalent destructive radio assay. The principles provided in this guide should be used to determine which type of measurement is best suited to the measurement application. This determination involves consideration of the characteristics of the items to be measured, as well as the goals of the measurement program. This guide applies to the suite of NDA instruments and measurement methods. It is incumbent upon the user to seek additional guidance within ASTM method-specific standards, as this guide does not take precedence. This guide may be applied to many situations spanning the range of nuclear materials from product through waste. Typical applications include: the measurement and characterization of transuranic wastes, low-level wastes, and mixed wastes; the determination of radioactivity below some regulatory threshold; scaling for non detected radionuclides, the measurement of safeguarded nuclear materials; shipper receiver confirmation; confirmation of nuclear material inventory; support of nuclear criticality safety evaluations; measurement of holdup of special nuclear material in process systems; support of decontamination and decommissioning activities; and in-situ analyzes of facilities, glove-boxes, hot cells, and the environment prior to and following demolition. This guide addresses elements of good practice such as; nuclear measurement instrumentation and its care; common hazards; ancillary personnel functions; facility readiness and requirements to support the NDA equipment; project scoping, requirements and objectives; assembly and deployment of the instrument;

calibration and test; computational modeling to augment physical testing; measurement validation; preventive maintenance; and the measurement control program

- (Draft Standard) “Standard Test Method for Nondestructive Assay by Tomographic Gamma Scanning”

**Scope:** This test method describes the nondestructive assay (NDA) of gamma ray emitting materials inside containers using tomographic gamma scanning (TGS). High resolution gamma ray spectroscopy is used to detect and measure the radionuclides of interest and to measure gamma ray attenuation from an external transmission source. The TGS method is most commonly used to quantify special nuclear material (SNM) enclosed in cylindrical containers. Typically cans or drums have been assayed up to 416L (110 gal) drums. Other sample shapes may be assayed but are constrained to the dimensions of the 416L drum. It has been successfully used to quantify  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{235}\text{U}$ . SNM loadings from one-tenth to several hundred grams of  $^{239}\text{Pu}$  and  $^{235}\text{U}$ . The TGS method is applicable to a wide range of item sizes and to a wide range of matrix material, with areal densities ranging from nearly zero (i.e., light combustibles) to more than  $1000 \text{ kg} \cdot \text{m}^{-3}$  (i.e., cemented sludge in 200 l drums). It is particularly well suited for items that have heterogeneous matrix materials and radionuclide distributions. Measured transmission values must be available to permit valid attenuation corrections. The particles containing the nuclides of interest must be small to minimize self-absorption of emitted gamma radiation. If not, then correction by calibration or other means may be required to avoid biased results. The mixture of material within each item voxel must be sufficiently uniform to apply an attenuation correction factor, computed from a measurement of gamma ray transmission through the voxel.

**Summary:** The assay of the nuclides of interest is accomplished by measuring the intensity of one or more characteristic gamma rays from each nuclide utilizing tomographic gamma scanning techniques. Corrections are made for count rate-related losses and attenuation by the item. The appropriate mass or efficiency calibration then provides the relationship between observed gamma ray intensity and nuclide content. Either of two distinct calibration methods can be used: 1. Isotope Specific (Mass) Calibration provides assay results for only those radionuclides for which the TGS is specifically calibrated. Calibration is performed using standards containing the radionuclides to be assayed. 2. Efficiency Curve Calibration entails determination of the system detection efficiency as a function of gamma ray energy. Analysis of assay data consists of using the energy of a peak to infer the emitting radionuclide, and then calculating the radionuclide mass from the specific activity and the gamma emission intensity of the radionuclide, and the corrected count rate and detector efficiency at the peak energy. The assay item is rotated about its vertical axis. Concurrently, the relative position of the assay item and detector are translated. This is repeated for every vertical segment. During this process, a series of measurements (grabs) are taken of gamma rays corresponding to radioisotopes of the transmission source and the emission source. Transmission and emission gamma ray measurements may be made concurrently or in series. From the transmission measurements, a 3-D map of the average linear attenuation coefficient of each voxel is determined. From the emission measurements, a 3-D map of the location of the gamma emitting radionuclides is determined. Through a voxel by voxel application of Beer’s law, the emission source strength is corrected for the attenuation of the matrix material. Count rate-dependent losses from pulse pile-up and analyzer deadtime are monitored and corrected for by electronic modules and/or

radioactive sources. TGS determines an estimate of the average attenuation coefficient of a voxel by measuring the attenuated intensity of an external transmission source at many different angular positions of the container. A collimator is used in front of the detector to restrict the measurement to a well-defined area. This also serves to reduce the background measured. The gamma ray spectra are measured using a high-resolution gamma detector (such as high-purity germanium). Two conditions must be met to optimize TGS assay results as follows the particles containing the nuclides of interest must be small enough to minimize self-absorption of emitted gamma radiation and The mixture of material within each item voxel must be uniform to apply an attenuation correction factor, generally computed from a measurement of gamma ray transmission through the voxel. If this condition is not met, then the result may be biased.

**Significance and use:** TGS provides a nondestructive means of measuring the nuclide content of items where the specific natures of the matrix and the chemical form and the relationship between the nuclide and matrix may be unknown. The procedure can serve as a diagnostic tool that provides 3-dimensional map of the transmission and nuclide concentration within an item. Sample preparation is limited to avoiding large quantities of heavily attenuating materials (such as lead shielding) in order to allow sufficient transmission through the container.

— (Very Early Draft Guide) “Standard Guide for Passive Gamma Measurements using Modeling”

**Scope:** This guide addresses modeling methods used to prepare for and to perform the nondestructive assay (NDA) of items containing radionuclides using passive gamma ray measurements. This guide includes information useful for general purpose in-situ measurements of items in a variety of configurations and good practices for the modeling of such configurations. In such cases, it is generally not practical to develop standards containing nuclear and matrix material that exactly match the characteristics of the sample in situ measurements. Mathematical modeling is used to estimate the response in such cases to produce a more accurate assay of the nuclear material of interest than a direct measurement based on calibrations for standards that do not match the configuration of the specific assay situation. A scientific knowledge of radiation sources and detectors, calibration procedures, geometry and error analysis is required for the users of this standard. This standard assumes that the user has, at a minimum, a basic understanding of these principles and good NDA practices, as defined for an NDA professional in ASTM C1490. The user of this standard also should have a basic understanding of the software used for modeling.

**Summary:** In-situ measurements may be significantly different than the measurement of other items measured in a laboratory environment. Frequently the quantity and distribution of nuclear material within a container is unknown. This guide provides methods of instrument calibration and determination of matrix attenuation corrections through modeling that may be used for the in-situ assay of radionuclides. The modeled correction factors are used to derive the activity of radionuclides in a container or configuration. Process knowledge and other records are used to obtain an estimate of the nuclides present in an item. Nuclide libraries are prepared to measure the expected nuclides, however this does not preclude the determination of other unexpected

nuclides. Process knowledge is also used to obtain preliminary information on the matrix materials that the analyst should expect in the container or location. This matrix information can be used in the modeling effort to determine the appropriate correction factors. Converting the measured data to estimates of the radionuclide activity requires careful evaluation of the measurement against the calibration assumptions, as well as a validation that any assumptions based on process knowledge were correct or of negligible impact on the assay result following model based correction of the count rates. Depending on the calibration, geometry, and measurement methods, corrections may be necessary for geometric effects (differences between activity measurement and calibration geometry), gamma ray attenuation effects (appropriate to the quantitative method chosen, for example, the container, item matrix, and absorbers), background, and interferences. Measurement uncertainties are estimated based on uncertainties in assay parameters, for example, activity distribution, attenuation effects, geometric effects, and measured count rates. In some cases, scans may be performed using low-resolution gamma ray detectors (e.g. NaI) to identify the location of activity within a container or the location and approximate concentration of a nuclide that may have contaminated soil, floors or walls. This data is used in the establishment of mathematical models to describe the physical location of equipment and materials. Typically, high purity germanium (HPGe) gamma ray spectrometers are then used to quantify the radioactivity. Corrections for attenuation and isotopic composition are included in this determination.

**Significance and use:** The following methods assist in demonstrating regulatory compliance in such areas as safeguards SNM inventory control, criticality control, decontamination and decommissioning, waste disposal and shipping. This guide can apply to the measurement of activity in equipment or containers, whose gamma ray attenuation properties can be measured or estimated by modeling, for which certified standards are not available. It may also be applied to in-situ measurements or to laboratory measurements for non-standard geometry samples. The guide may also be applied to spectra obtained from high resolution or low resolution gamma ray detectors.

#### ASTM Standards under the Jurisdiction of C26.13 Spent Fuel and High Level Waste

- C1174-97 “Standard Practice for Prediction of the Long-Term Behaviour of Materials, Including Waste Forms, Used in Engineered Barrier Systems (EBS) for Geological Disposal of High-Level Radioactive Waste”

This standard covers steps for the development of methods to aid in the prediction of the long-term behaviour of materials, such as “engineered barrier” system (EBS) materials and waste forms, used in the geologic disposal of high-level nuclear waste. These steps include problem definition, testing, modeling, and confirmation. The predictions are based on models derived from interpretation of data obtained from tests and appropriate analogs. The purpose of this practice is to provide information to serve as part of the basis for performance assessment of a geologic repository. This standard is intended to guide in making predictions of alterations in materials over periods of time beyond which empirical data can be used for the accurate assessment of performance and behaviour.

C1285-02 “Standard Test Methods for Determining Chemical Durability of Nuclear, Hazardous, and Mixed Waste Glasses and Multiphase Glass Ceramics: The Product Consistency Test (PCT)”

This test evaluates the chemical durability of homogeneous glasses, phase separated glasses, devitrified glasses, glass ceramics, and/or multiphase glass ceramic waste forms collectively referred to as “glass waste forms” by measuring the concentrations of the chemical species released to a test solution. There are two test methods, A and B. Test Method A is a seven-day chemical durability test performed at  $90 \pm 2^\circ\text{C}$  in a leachant of deionized water. In this test method the glass waste form is crushed and sieved to a size range of 0.149–0.074 mm. The test method is static and conducted in stainless steel vessels. Test Method A can specifically be used to evaluate whether the chemical durability and elemental release characteristics of nuclear, hazardous, and mixed glass waste forms have been consistently controlled during production. Test Method B is also a static chemical durability test but allows for testing at various test durations, test temperatures, mesh size, mass of sample, leachant volume, and leachant compositions. These test methods provide data useful for evaluating the chemical durability of glass waste forms as measured by elemental release. Accordingly, they may be applicable throughout manufacturing, research, and development. Test Method B can specifically be used to measure the chemical durability of glass waste forms under various leaching conditions, for example, varying test durations, test temperatures, ratio of sample-surface area to leachant volume, and leachant types. Data from this test may form part of the larger body of data that are necessary in the logical approach to long-term prediction of waste form behavior (see ASTM C1174).

— C1431-99 “Standard Guide for Corrosion Testing of Aluminum-Based Spent Nuclear Fuel in Support of Repository Disposal”

This guide covers corrosion testing of aluminum-based spent nuclear fuel in support of geologic repository disposal (per the requirements in 10 CFR 60 and 40 CFR 191). The testing described in this publication is designed to provide data for analysis of the chemical stability and radionuclide release behavior of aluminum-based waste forms produced from aluminum-based spent nuclear fuels. The data and analyzes from the corrosion testing will support the technical basis for inclusion of aluminum-based spent nuclear fuels in the repository source term. Interim storage and transportation of the spent fuel will precede geologic disposal; therefore, reference is also made to the requirements for interim storage (per 10 CFR 72) and transportation (per 10 CFR 71). The analyses that will be based on the data developed are also necessary to support the safety analyzes reports (SARs) and performance assessments (PAs) for disposal systems.

Spent nuclear fuel that is not reprocessed must be safely managed prior to transportation to, and disposal in, a geologic repository. Placement in an interim storage facility may include direct placement of the irradiated fuel or treatment of the fuel prior to placement, or both. The aluminum-based waste forms may be required to be ready for geologic disposal, or road ready, prior to placement in extended interim storage. Interim storage facilities, in the United States, handle fuel from civilian commercial power reactors, defense nuclear materials production reactors, and research reactors. The research reactors include both foreign and domestic reactors. The aluminum-based fuels

in the spent fuel inventory in the United States are primarily from defense reactors and from foreign and domestic research reactors. The aluminum-based spent fuel inventory includes several different fuel forms and levels of <sup>235</sup>U enrichment. Highly enriched fuels (<sup>235</sup>U enrichment levels > 20%) are part of this inventory.

Knowledge of the corrosion behaviour of aluminum-based spent nuclear fuels is required to ensure safety and to support licensing or other approval activities, or both, necessary for disposal in a geologic repository. The response for the aluminum-based spent nuclear fuel waste form(s) to disposal environments must be established for configuration-safety analyzes, criticality analyzes, PAs, and other analyzes required to assess storage, treatment, transportation, and disposal of spent nuclear fuels. This is particularly important for the highly enriched, aluminum-based spent nuclear fuels. The test protocols described in this guide are designed to establish material response under the repository relevant conditions.

The majority of the aluminum-based spent nuclear fuels are aluminum clad, aluminum-uranium alloys. The aluminum-uranium alloy typically consists of uranium aluminide particles dispersed in an aluminum matrix. Other aluminum-based fuels include dispersions of uranium oxide, uranium silicide, or uranium carbide particles in an aluminum matrix. These particles, including the aluminides, are generally cathodic to the aluminum matrix. Selective leaching of the aluminum in the exposure environment may provide a mechanism for redistribution and relocation of the uranium-rich particles. Particle redistribution tendencies will depend on the nature of the aluminum corrosion processes and the size, shape, distribution and relative reactivity of the uranium-rich particles. Interpretation of test data will require an understanding of the material behaviour. This understanding will enable evaluation of the design and configuration of the waste package to ensure that unfilled regions in the waste package do not provide sites for the relocation of the uranium-rich particles into nuclear critical configurations. Test samples must be evaluated, prior to testing, to ensure that the size and shape of the uranium-rich particles in the test samples are representative of the particles in the waste form being evaluated.

The use of the data obtained by the testing described in this guide will be optimized to the extent the samples mimic the condition of the waste form during actual repository exposure. The use of Practice C 1174 is recommended for guidance. The selection of test samples, which may be unaged or artificially aged, should ensure that the test samples and conditions bound the waste form/repository conditions. The test procedures should carefully describe any artificial aging treatment used in the test program and explain why that treatment was selected.

— C1454-00 Standard Guide for Pyrophoricity/Combustibility Testing in Support of Pyrophoricity Analyzes of Metallic Uranium Spent Nuclear Fuel

This guide describes testing protocols for pyrophoricity, or combustibility characteristics, or both, of metallic uranium-based SNF. The testing will provide basic data for input into more detailed computer codes or analyzes of thermal, chemical, and mechanical SNF responses. These analyzes would support the engineered barrier system (EBS) design bases and safety assessment of extended interim storage facilities and final disposal in a geologic repository. The testing also could provide data related to licensing

requirements for the design and operation of a monitored retrievable storage facility (MRS) or independent spent fuel storage installation (ISFSI).

This guide describes testing of metallic uranium spent nuclear fuel (SNF) in support of transportation (in accordance with the requirements of 10CFR71), interim storage (in accordance with the requirements of 10CFR72), and geologic repository disposal (in accordance with the requirements of 10CFR60). The testing described herein is designed to provide basic data related to the evaluation of the pyrophoricity/combustibility characteristics of containers or waste packages containing metallic uranium SNF in support of safety analyzes (SAR), or performance assessments (PA) of transport, storage, or disposal systems, or a combination thereof.

Spent nuclear fuel that is not reprocessed must be emplaced in secure temporary interim storage as a step towards its final disposal in a geologic repository. In the United States, SNF, from both civilian commercial power reactors and defense nuclear materials production reactors, will be sent to interim storage, and subsequently, to deep geologic disposal. U.S. commercial SNF comes predominantly from light water reactors (LWRs) and is uranium dioxide-based, whereas U.S. Department of Energy (DOE) owned defense reactor SNF is in several different chemical forms, but is predominantly (80 % by weight of uranium) metallic uranium-based.

Knowledge of the pyrophoricity/combustibility characteristics of the SNF is required to support licensing activities for extended interim storage and ultimate disposition in a geologic repository. These activities could include interim storage configuration safety analyzes, conditioning treatment development, preclosure design basis event (DBE) analyzes of the repository controlled area, and postclosure performance assessment of the EBS.

Metallic uranium fuels are clad, generally with zirconium, aluminum, stainless steel, or magnesium alloy, to prevent corrosion of the fuel and to contain fission products. If the cladding is damaged and the metallic SNF is stored in water the consequent corrosion and swelling of the exposed uranium may enhance the chemical reactivity of the SNF by further rupturing the cladding and creating uranium hydride particulates and/or inclusions. The condition of the metallic SNF will affect its behaviour in transport, interim storage or repository emplacement, or both, and therefore, influence the engineering decisions in designing the pathway to disposal.

The interpretation of the test data depends on the characteristics of the sample tested. The type and the size of the SNF sample must be chosen carefully and accounted for in the usage of the data. The use of the data obtained by the testing described herein may require that samples be used which mimic the condition of the SNF at times far into the future, for example, the repository post containment period. This guide does not specifically address methods for 'aging' samples for this purpose. The section in Practice C 1174 concerning the accelerated testing of waste package materials is recommended for guidance on this subject.

- C1562-03 “Standard Guide for Evaluation of Materials Used in Extended Service of Interim Spent Nuclear Fuel Dry Storage Systems”

Part of the total inventory of commercial spent nuclear fuel (SNF) is stored in dry cask storage systems (DCSS) under licenses granted by the U.S. Nuclear Regulatory Commission (NRC). The purpose of this guide is to provide information to assist in supporting the renewal of these licenses, safely and without removal of the SNF from its licensed confinement, for periods beyond those governed by the term of the original license. This guide provides information on materials behaviour under conditions that may be important to safety evaluations for the extended service of the renewal period. This guide is written for DCSS containing light water reactor (LWR) fuel that is clad in zirconium alloy material and stored in accordance with the Code of Federal Regulations (CFR), at an independent spent-fuel storage installation (ISFSI). The components of an ISFSI, addressed in this publication, include the commercial SNF, canister, cask, and all parts of the storage installation including the ISFSI pad. The language of this guide is based, in part, on the requirements for a dry SNF storage license that is granted, by the U.S. Nuclear Regulatory Commission (NRC), for up to 20 years. Although government regulations may differ for various nations, the guidance on materials properties and behaviour given here is expected to have broad applicability.

This guide addresses many of the factors affecting the time-dependent behaviour of materials under ISFSI service [10 CFR Part 72.42]. These factors are those regarded to be important to performance, in license extension, beyond the currently licensed 20-year period. Examples of these factors are given in this guide and they include materials alterations or environmental conditions for components of an ISFSI system that, over time, could have significance related to safety. For purposes of this guide, a license period of an additional 20 to 80 years is assumed.

This guide addresses the determination of the conditions of the spent fuel and storage cask materials at the end of the initial 20-year license period as the result of normal events and conditions. However, the guide also addresses the analysis of potential spent fuel and cask materials degradation as the result of off-normal, and accident-level events and conditions that may occur during any period.

This guide provides information on materials behaviour to support continuing compliance with the safety criteria, which are part of the regulatory basis, for licensed storage of SNF at an ISFSI. The safety functions addressed and discussed in this standard guide include thermal performance, radiological protection, confinement, sub-criticality, and retrievability. The regulatory basis includes 10 CFR Part 72 and supporting regulatory guides of the U.S. Nuclear Regulatory Commission. The requirements set forth in these publications indicate that the following items were considered in the original licensing decisions: properties of materials, design considerations for normal and off-normal service, operational and natural events, and the bases for the original calculations. These items may require reconsideration of the safety-related arguments that demonstrate how the systems continue to satisfy the regulatory requirements. Further, to ensure continued safe operation, the performance of materials must be justified in relation to the effects of time, temperature, radiation field, and environmental conditions of normal and off-normal service. Arguments for long-term performance must account for materials alterations (especially degradations) that are expected during the service periods, which include the periods of the initial license and of the license renewal. This guide pertains only to structures, systems, and components important to safety during extended storage period and during retrieval functions, including transport and transfer operations. Materials information that

pertains to safety functions, including retrieval functions, is pertinent to current regulations and to license renewal process, and this information is the focus of the guide. This guide is not intended to supplant the existing regulatory process.

Standards currently under development by ASTM C26.13:

- DRAFT WK82 “Standard Practice for Measurement of the Glass Dissolution Rate Using the Single-Pass Flow-Through Test Method”

This standard describes a single-pass flow-through (SPFT) test method that can be used to measure the dissolution rate of a homogeneous silicate glass, including nuclear waste glasses, in various test solutions at temperatures less than 100°C. Tests may be conducted under conditions in which the effects from dissolved species on the dissolution rate are minimized to measure the forward dissolution rate at specific values of temperature and pH, or to measure the dependence of the dissolution rate on the concentrations of various solute species. Tests are conducted by pumping solutions in either a continuous or pulsed flow mode through a reaction cell that contains the test specimen. Tests must be conducted at several solution flow rates to evaluate the effect of the flow rate on the glass dissolution rate. This standard excludes static test methods in which flow is simulated by manually removing solution from the reaction cell and replacing it with fresh solution. Tests may be conducted with demineralized water, chemical solutions (such as pH buffer solutions, simulated groundwater solutions, and brines), or actual groundwater. Tests may be conducted with crushed glass of a known size fraction or monolithic specimens having known geometric surface area. The reacted solids may be examined to provide additional information regarding the behavior of the material in the test and the reaction mechanism. Tests may be conducted with glasses containing radionuclides. Data from these tests can be used to determine the values of kinetic model parameters needed to calculate the glass corrosion behavior in a disposal system over long times. This SPFT test method can be used to characterize various aspects of glass corrosion behavior that can be utilized in a mechanistic model for calculating long-term behavior of a nuclear waste glass. In addition, depending on the values of test parameters that are used, the results of SPFT tests can be used to measure the intrinsic dissolution rate of a glass, the temperature and pH dependencies of the rate, and the effects of various dissolved species on the corrosion behavior.

- WK84 “Standard Test Method for Measuring Waste Glass Durability by Vapor Hydration Test”

The vapor hydration test (VHT) method can be used to study durability by measuring the corrosion of a waste forms such as glasses and glass ceramics upon exposure to water vapor at elevated temperatures. The abundance of alteration phases that form can be used as a measure of the extent of corrosion. These tests provide useful information regarding the alteration phases that are formed, the disposition of radioactive and hazardous components, and the alteration kinetics under the specific test conditions. This information can be used in site performance assessment evaluations. For the vapor hydration tests, glass specimens are suspended from a support rod with platinum wire within a sealed test vessel. A volume of water determined by the volume of the test vessel and the test temperature is added to the vessel. The vessel is then sealed and placed in an oven at the desired test temperature and left undisturbed. After the desired test duration, the vessel is removed from the oven and the bottom of the vessel is cooled

to condense the vapor in the vessel. Test samples are removed and examined with optical microscopy, XRD, SEM, and other analytical methods. The remaining glass thickness is measured and alteration phases are identified. The Vapor Hydration Test can be used to study the corrosion of waste forms under conditions of high temperature and contact by water vapor or thin films of water. This method may serve as an accelerated test for some materials, since the high temperatures will accelerate thermally activated processes. It can be used as a screening test to determine the propensity of waste glasses to alter and for relative comparison between waste forms.

- DRAFT Z7529Z “Standard Test Methods for Determining the Liquidus Temperature ( $T_L$ ) of Immobilized Waste Glasses and Simulated Waste Glasses”

The standard is used to measure the liquidus temperature ( $T_L$ ) of immobilized waste glasses or simulated waste glasses in the temperature range of approximately 600 to 1500°C. Here "waste glass" refers to a glass used to immobilize radioactive, mixed, or hazardous wastes. Two methods are used in the standard. The gradient temperature (GT) method should be used only for glasses with low volatility near  $T_L$ . The uniform temperature (UT) method can be used for glasses with higher volatility near  $T_L$  and is more suitable for measurements that are made remotely. The GT and UT methods address concerns specific to the measurement of  $T_L$  in glasses with low viscosity and potential high volatility near  $T_L$ , such as waste glasses. These methods are valid for glasses that do not crystallize during normal quenching on stainless steel pour plate from  $T_L$  to room temperature. Reproducible measurement of  $T_L$  is useful in waste glass plant operation, glass formulation, and melter design. Knowledge of  $T_L$  provides for assessing the minimum temperature that must be maintained in a waste glass melt to ensure that crystallization does not occur. Information about  $T_L$  is also important in glass formulation to develop glasses compatible with planned melter technology.

- DRAFT Z8671Z “Standard Test Methods for Determining the Amount of Devitrification in a Nuclear Waste Glass and for Constructing Time-Temperature-Transformation (TTT) Diagrams”

These time-temperature-transformation (TTT) test methods evaluate the type and amount of devitrification in heat treated or canistered nuclear or mixed waste glass. Test Method A describes the methods by which qualitative TTT diagrams that delineate crystallizing phase regions can be determined for heat treated or canistered nuclear, mixed, or hazardous waste glasses. Test Method B describes the methods by which quantitative TTT diagrams can be determined for nuclear, mixed, or hazardous waste glasses. The quantitative TTT diagrams delineate crystallizing phase regions by determination of the volume or weight percent crystallinity. The quantitative TTT diagrams and the associated kinetic analysis developed using Test Method B can specifically be used to evaluate the degree to which a glass will crystallize when subjected to varying time-temperature conditions.

- DRAFT WK80 “Standard Guide for Drying Behaviour of Spent Nuclear Fuel”

This standard guide is organized to discuss the three major components of significance in the drying behaviour of spent nuclear fuel: evaluating the need for drying, drying spent nuclear fuel, and confirmation of adequate dryness.

The guide addresses drying methods and their limitations in drying spent nuclear fuels that have been in storage at water pools. The sources and forms of water that may remain in SNF and/or its container after the drying process and discusses the importance and potential effects they may have on fuel integrity, and container materials. The effects of residual water are discussed mechanistically as a function of the container thermal and radiological environment to provide guidance on situations that may require extraordinary drying methods, specialized handling, or other treatments.

The basic issue in drying is to determine how dry the SNF must be in order to prevent issues with fuel retrievability, container pressurization, or container corrosion. Adequate dryness may be readily achieved for undamaged commercial fuel but may become a complex issue for any SNF where cladding damage has occurred during fuel irradiation and/or storage at the spent fuel pools. Dryness issues may also result from the presence of sludge, crud, and other hydrated compounds connected to the SNF that hold water and resist drying efforts.

This guide does not purport to address all the safety concerns, if any, associated with its use. It is the responsibility of the user of this guide to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.

- DRAFT WK83 “Standard Guide for the Characterization of Spent Nuclear Fuel in Support of Geologic Repository Disposal”

This Standard Guide provides guidance for the types and extent of testing that would be involved in characterizing the physical and chemical nature of spent nuclear fuel (SNF) in support of analyzes of its disposal in a geologic repository. The testing is designed to provide information that supports the design, safety analysis, and performance assessment of a geologic repository for the ultimate disposal of the SNF. The testing described includes characterization of such physical attributes as physical appearance, weight, density, shape/geometry, and degree and type of cladding damage. The testing described also includes the measurement/examination of such chemical attributes as radionuclide content, microstructure, and corrosion product content, and such environmental response characteristics as drying rates, oxidation rates (in dry air, water vapor, and liquid water), ignition temperature, and dissolution/degradation rates. It is assumed in formulating the SNF characterization activities in this publication that the SNF has resided in an interim storage facility, either wet (e.g., a spent fuel pool) or dry (e.g., an independent spent fuel storage facility or ISFSI) in the time between reactor discharge and being sent to the geologic repository, and that the manner of interim storage may affect the SNF characteristics.

- DRAFT “Standard Test Method for Measuring the Dissolution Rate of Spent Nuclear Fuel in Dilute Aqueous Solutions Using a Flowthrough Technique”

This test method was written specifically for measuring the dissolution rates of  $\text{UO}_2$  and mixed oxide ( $\text{UO}_2/\text{PuO}_2$ , referred to as MOX) spent nuclear fuel (SNF) specimens and corresponding unirradiated fuel specimens. The purpose of this test method is to provide dissolution rate data for use in the assessment of SNF performance in a geologic repository. The test is conducted under conditions where the concentrations of the dissolving uranium and other elements in the test solution remain well below their solubility limits thereby precluding any back reactions or reactions involving

precipitation of secondary phases. This enables unambiguous measurement of the forward reaction rate. In addition, the test results can be used to determine whether the SNF dissolves congruently or, instead, if certain radionuclides or phases dissolve preferentially.

This test is limited to unirradiated  $\text{UO}_2$  and  $\text{UO}_2$  or MOX spent fuel (plus unirradiated  $\text{UO}_2$  or MOX fuel) in the form of powder or fragments. This test is considered to be a Characterization Test in the context of ASTM Standard Practice C-1174 only when the surface area that is actually contacted by the water is determined for the test specimen. In that case, this method provides a measure of the fundamental reaction kinetics between the fuel and the particular aqueous solution being used in the test. The dissolution rate measured in a Characterization Test is also known as the intrinsic specific dissolution rate of the material under the conditions of the test. It is important to note that  $\text{UO}_2$  spent fuel test specimens that consist of multigrain fragments (typically 1 to 5 mm in size) generally cannot be used to measure intrinsic specific dissolution rates because the surface area contacted by water cannot be determined by any known method. Fragment specimens of  $\text{UO}_2$  spent fuel can be tested by this method, but the data apply only to the particular specimen tested and, in particular, do not provide an intrinsic specific dissolution rate.

#### ASTM Standards under the Jurisdiction of C26.14 Remote Systems

- C1533-02 “Standard Guide for General Design Considerations for Hot Cell Equipment”

*Scope:* The intent of this guide is to provide general design and operating considerations for the safe and dependable operation of remotely operated hot cell equipment. Hot cell equipment is hardware used to handle, process, or analyze nuclear or radioactive material in a shielded room. The equipment is placed behind radiation shield walls and cannot be directly accessed by the operators or by maintenance personnel because of the radiation exposure hazards. Therefore, the equipment is operated remotely, either with or without the aid of viewing.

This guide may apply to equipment in other radioactive remotely operated facilities such as suited entry repair areas, canyons or caves, but does not apply to equipment used in commercial power reactors.

This guide does not apply to equipment used in gloveboxes.

- C1533-02 "Standard Guide for Materials Handling Equipment for Hot Cells"

*Scope:* This guide covers materials handling equipment used in hot cells (shielded cells) for the processing and handling of nuclear and radioactive materials. The intent of this guide is to aid in the selection and design of materials handling equipment for hot cells in order to minimize equipment failures and maximize the equipment utility.

It is intended that this guide record the principles and caveats that experience has shown to be essential to the design, fabrication, installation, maintenance, repair, replacement, and, decontamination and decommissioning of materials handling equipment capable of

meeting the stringent demands of operating, dependably and safely, in a hot cell environment where operator visibility is limited due to the radiation exposure hazards.

This guide may apply to materials handling equipment in other radioactive remotely operated facilities such as suited entry repair areas and canyons, but does not apply to materials handling equipment used in commercial power reactors.

- C1572-04 "Standard Practice/Guide for Dry Lead Glass and Oil-Filled Lead Glass Radiation Shielding Window Components for Remotely Operated Facilities"

*Scope:* This standard establishes the minimum requirements for the design, fabrication, quality assurance, inspection, testing, packaging, shipping, installation and maintenance of radiation shielding window components. These window components include wall liner embedments, dry lead glass radiation shielding window assemblies, oil-filled lead glass radiation shielding window assemblies, shielding wall plugs, barrier shields, view ports, and the installation/extraction table required for the installation and operation of the window components.

This standard is intended for those persons who are tasked with the planning, design, procurement, fabrication, installation, and operation of the radiation shielding window components that may be used in the operation of hot cells, high level caves, mini-cells, canyon facilities, and very high level radiation areas.

This standard applies to radiation shielding window assemblies used in normal concrete walls, high-density concrete walls, steel walls and lead walls.

#### Standards currently under development by ASTM C26.14

- DRAFT Z9040Z "Standard Guide for Mechanical Drive Systems for Remote Operation in Hot Cell Facilities"

*Scope:* The intent of this guide is to provide general guidelines for the design, selection, and strategies for operations, maintenance, and recovery of mechanical drive systems used in remote hot cell environments. The term mechanical drive systems used herein, encompasses all individual components used for imparting motion to equipment systems, subsystems, assemblies, and other components. It also includes complete positioning systems and individual units that provide motive power and any position indicators necessary to monitor the motion.

This guide is intended to be applicable to equipment that cannot be accessed directly for purposes of operation or maintenance, nor can the equipment be viewed directly, for example, without shielded viewing windows, periscopes, or a video monitoring system.

- DRAFT Z8524Z "Standard Practice/Guide for Viewing Systems for Remotely Operated Facilities"

*Scope:* This guide covers viewing systems for remotely operated facilities, including hot cells (shielded cells), used for the processing and handling of nuclear and radioactive materials. The intent of this guide is to aid in the selection and design of remote viewing systems to maximize their usefulness and to minimize equipment failures.

This guide does not cover shielding windows used for hot cell viewing.

It is intended that this guide record the principles and caveats that experience has shown to be essential to the design, fabrication, installation, maintenance, repair, replacement, and, decontamination and decommissioning of remote viewing equipment capable of meeting the stringent demands of operating, dependably and safely, in a hot cell environment where operator visibility is limited due to the radiation exposure hazards.

This guide is intended to apply to applications of remote viewing for nuclear applications but may be applicable to any environment where remote operational viewing is desirable.

**Note:** Department of Energy (DOE) publications from 1995 forward can be found at the Office of Scientific and Technical Information (OSTI) Web site “Information Bridge” <http://www.osti.gov/bridge/> . The Information Bridge” provides an open source to full-text and bibliographic records of DOE research and development reports.

- S.C. Goheen, M. McCulloch, B.L. Thomas, R.G. Riley, D.S. Sklarew, G.M. Mong, and S.K. Fadeff, “DOE Methods for Evaluating Environmental and Waste Management Samples,” Publication number DOE/EM-0089T, Rev. 2

**Abstract:**

DOE Methods for Evaluating Environmental and Waste Management Samples (DOE Methods) is a resource intended to support sampling and analytical activities for the evaluation of environmental and waste management samples from U.S. Department of Energy (DOE) sites. DOE Methods is the result of extensive cooperation from all DOE analytical laboratories. DOE Methods is designed to encompass methods for collecting representative samples and for determining the radioisotope activity and organic and inorganic composition of a sample. These determinations will aid in defining the type and breadth of contamination and thus determine the extent of environmental restoration or waste management actions needed, as defined by the DOE, the U.S. Environmental Protection Agency, or others. The development of DOE Methods is supported by the Analytical Services Division of DOE. Methods appearing in this publication are one of two types, “Draft” or “Verified”. “Draft” methods that have been reviewed internally and show potential for eventual verification are included in this publication, but they have not been reviewed externally, and their precision and bias may not be known. “Verified” methods in DOE Methods have been reviewed by volunteers from various DOE sites and private corporations. These methods have delineated measures of precision and accuracy. The publication contains quality assurance (QA), quality control (QC), safety, sampling, organic analysis, inorganic analysis, and radio-analytical guidance as well as sampling and analytical methods.

- DOE Standard, DRAFT Destructive Assay Methods Compendium, Publication No. DOE-STD-XXX-Year, U.S. Department of Energy, Washington, D.C. 20584 Draft dated September 27, 2002

This standard covers destructive analysis analytical chemistry methods which are useful in nuclear material control and accounting (MC&A). A large part of Special Nuclear Material (SNM) inventories and virtually all of the highly attractive SNM inventories are based on sampling bulk materials followed by destructive assay (DA) of these materials. The measurement methods described in the publication support MC&A in process control, physical inventory verification, evaluation of the effects of process changes, detecting and resolving shipper-receiver differences, and the resolution of IAEA inspector-facility differences. The evaluation and the specification of DA, MC&A techniques have proven difficult, in part, because of the highly specialized and technical nature of DA and because of the wide variety of methods and applications. The destructive assay compendium is provided as a resource for those selecting methods for use and for those evaluating the performance of MC&A measurements. The compendium is limited to analytical methods used to quantify and characterize plutonium (Pu) and enriched uranium (EU).

- Toxicity Characteristic Leaching Procedure (TCLP). 1992. SW-846, Method 1311. Rev. 2. In *Test Methods for Evaluating Solid Waste, Volume 1C: Laboratory Manual Physical/Chemical Methods*, U.S. Environmental Protection Agency, Office of Solid Waste and Emergency Response, Washington, D.C.

This leaching procedure is designed to determine the mobility of both inorganic and organic analytes present in liquid, solid, and combined liquid/solid wastes. The TCLP is conducted to determine if the waste should have a dangerous waste designation. TCLP “extracts” are prepared from both liquid and solid wastes for analysis. Liquid wastes are filtered through a 0.6 to 0.8  $\mu\text{m}$  glass fiber filter to generate the TCLP extract. For solid waste samples, particle size reduction may be required as the particles must be capable of passing through a 9.5-mm (0.375-in.) standard sieve. The reduced solid, a minimum 100 g sample is recommended, is placed into an extractor vessel with an extraction fluid equal to 20 times the weight of the solid sample. The sample is rotated head over heel at 30 rpm for 18 h at ambient temperature. The liquid is then separated from the solid phase by filtration, and the leachate is then analyzed for the analytes of concern. For liquid/solid composite waste samples, the liquid is filtered from the solid phase and stored for later analysis. If the initial liquid phase does not form an immiscible phase with the resultant solid TCLP liquid extract they are combined and analyzed together. If the two liquids form an immiscible mixture then they are analyzed separately and combined mathematically to yield a volume-weighted average concentration. A Zero-Headspace Extraction Vessel (ZHE) is used if the waste is being tested for the mobility of volatile analytes.



## Annex IV

### BIBLIOGRAPHY

This annex includes a compendium of references related to the different sections of the main publication. The intention of having this compendium is to provide the operators of the characterization laboratories with a large set of information is required. It deals with the different technical aspects of the publication such as statistics, analysis, radiochemistry etc.

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## Annex V

### EXAMPLE OF UNCERTAINTY CALCULATION FOR $^{90}\text{Sr}$ DETERMINED BY LSC AFTER CHROMATOGRAPHIC SEPARATION PROCESS

#### 1. HANDOUT FOR DEFINITIONS & SYMBOLS

**A measurement**

A set of operations having the object of determining a value of a quantity.

**The measurand**

The particular quantity subject to measurement

**True value**

A value that would be obtained by a perfect measurement

**Uncertainty of measurement**

A parameter associated with the result of a measurement, which characterises the spread of the values that could be reasonably attributed to the results.

**Type A uncertainty**

Evaluated by calculation using statistical methods and usually based on a series of repeat observations.

**Type B uncertainty**

Evaluated by means other than used for type A

**Error**

The result of a measurement minus the true value of the measurand

**Output quantity:  $y$** 

The final result of a measurement process

**Input quantity:  $x_i$** 

Quantities upon which the output quantity  $y$  depends.

**Standard uncertainty:  $u$** 

The uncertainty of the results of a measurement expressed as standard deviation

**Standard uncertainty  $u(x_i)$  associated with each input quantity  $x_i$** **a) For a type A uncertainty:**

$u(x_i)$  is the standard deviation, calculated by statistical or mathematical methods. It is usually calculated from a series of repeated observations, as the standard deviation of the mean value

**b) For a type B uncertainty:**

$u(x_i)$  is the estimated standard deviation, evaluated by means other than used for type A. The standard deviation will depend on assumed or guessed probability distribution.

- i. if the extreme limits ( $\pm'a'$ ) of the distribution are known or assumed, the standard uncertainty is given by the limit 'a' divided by a number associated with the assumed probability distribution as follows:

Distribution type	divisor
Rectangular (or uniform)	$\sqrt{3}$
Triangular	$\sqrt{6}$
U-shaped	$\sqrt{2}$

- ii. If a normal distribution is assumed, but a multiple of the standard uncertainty is obtained by divided the supplied uncertainty by that multiple

Distribution type	divisor
Normal (k=1)	1
Normal (k=2)	2
Normal (k=3)	3

### Arithmetic mean: $\bar{q}$

Average of n repeat observations -  $q_j$ , of randomly varying quantity q

$$\bar{q} = \frac{1}{n} \sum_{j=1}^n q_j$$

### Standard deviation (of a population)

a measure of the spread of a set of results, describing how values typically differ from the average of the set.

$$s(q_j) = \sqrt{\frac{\sum_{j=1}^n (q_j - \bar{q})^2}{n}}$$

For n approaching infinity

### Experimental standard deviation (of a population): $s(q_j)$

The estimate of the standard deviation of the whole 'population' but based on a limited sample from that population.

$$s(q_j) = \sqrt{\frac{\sum_{j=1}^n (q_j - \bar{q})^2}{n-1}}$$

### Experimental variance (of a population): $s^2(q_j)$

The estimate of the variance of the 'population', based on a limited is

$$s^2(q_j) = [s(q_j)]^2 = \frac{\sum_{j=1}^n (q_j - \bar{q})^2}{n-1}$$

### Standard deviation of the mean: $s(\bar{q})$

For a sample of n results, the uncertainty of the mean is:  $s(\bar{q}) = \frac{s(q_j)}{\sqrt{n}}$  where the uncertainty of a typical single result,  $s(q_j)$ , may be evaluated either

a) From the same set of  $n$  repeat measurements used to evaluate the mean, then

$$s(\bar{q}) = \sqrt{\frac{\sum_{j=1}^n (q_j - \bar{q})^2}{n(n-1)}}$$

or

b) From a separate set of  $n^*$  repeat measurements, then

$$s(q_j) = \sqrt{\frac{\sum_{i=1}^n (q_j^* - \bar{q}^*)^2}{n^* - 1}} \quad \text{and therefore} \quad s(\bar{q}) = \sqrt{\frac{\sum_{i=1}^n (q_j^* - \bar{q}^*)^2}{n(n^* - 1)}}$$

### Degrees of freedom: $\nu_i$

The number of terms in a sum minus the number of constraints on the terms of the sum.

### Degrees of freedom of the standard uncertainty $u(x_i)$ of an input estimate $x_i$ : $\nu_i$

Type A:  $\nu_i = n-1$  ( $n$ : number of measurements made to evaluate type A uncertainty)

Type B:  $\nu_i =$  can be regarded to infinity (usually)

### Sensitivity coefficient: $c_i$

The sensitivity coefficient for an input quantity  $x_i$  is a factor which relates a small change in  $x_i$  to the consequential small fractional change in output quantity  $y$ . If the relationship can be expressed as  $y = f(\dots x_i \dots)$  then:

$$c_i = \frac{\partial y}{\partial x_i}$$

### Sensitivity coefficient for relative values: $c_i^{\text{rel}}$

The sensitivity coefficient (for relative values) for an input quantity  $x_i$  is a factor, which relates a small change in  $x_i$  to the consequential small fractional change in output quantity  $y$ .

$$c_i^{\text{rel}} = (\partial y / y) / \partial x_i / x_i = c_i \cdot x_i / y$$

### Component of standard uncertainty of the output estimate $y$ , due to the uncertainty in $x_i$ : $u_i(y)$

Given by  $u_i(y) = c_i \cdot u(x_i)$ , since  $u_i(y)$  is only used as  $u_i^2(y)$ , it is usual to make  $u_i(y)$  positive, so the strict definition is:

$$u_i = |c_i| \cdot u(x_i)$$

### Law of propagation of uncertainties

When there are no correlated, the law of propagation of uncertainties states that when several uncertainty distribution are combined, the total variance,  $V$ . Of the combined uncertainty distribution will be the sum of the individual component variances  $\nu_i$ :  $V = \sum \nu_i$

Alternatively, in terms of standard uncertainties of  $y$  and of input parameters  $x_i$  and the sensitivity coefficients  $c_i$ , where  $V = u_c^2(y)$  and  $v_i = c_i^2 \cdot u_i^2(x_i) = u_i^2(y)$ . The law can be expressed, in terms of component uncertainties in  $y$ , as

$$u_c^2(y) = \sum u_i^2(y)$$

or in terms of component uncertainties in  $x_i$

$$u_c^2(y) = \sum c_i^2 \cdot u_i^2(x_i)$$

If any of the component quantities are correlated, there are extra covariance terms, which are induced by the generalised version of the Law,  $V = \sum v_i + \sum \sum \text{cov}(x_i, x_j)$ , which can be expressed as

$$u_c^2(y) = \sum c_i^2 \cdot u_i^2(x_i) + \sum \sum c_i c_j \text{cov}(x_i, x_j)$$

### Combined standard uncertainty of the output estimate $y$ : $u_c(y)$

Based on Law of Propagation of Uncertainties

When there are no correlations, the law of propagation of uncertainties states that when several uncertainty distribution are combined, the total variance,  $V$ . Of the combined uncertainty distribution will be the sum of the individual component variances  $v_i$ :  $V = \sum v_i$

Alternatively, in terms of standard uncertainties of  $y$  and of input parameters  $x_i$  and the sensitivity coefficients  $c_i$ , where  $V = u_c^2(y)$  and  $v_i = c_i^2 \cdot u_i^2(x_i) = u_i^2(y)$ . The law can be expressed, in terms of component uncertainties in  $y$ , as

$$u_c(y) = \sqrt{\sum_{i=1}^N u_i^2(y)} \quad \text{or} \quad \frac{u_c(y)}{y} = \sqrt{\sum_{i=1}^N \left( \frac{u_i(y)}{y} \right)^2} \quad \text{for the case of no correlations.}$$

In terms of the absolute or relative sensitivity coefficients and the uncertainty of the input quantities,

$$u_c(y) = \sqrt{\sum_{i=1}^N c_i^2 \cdot u^2(x_i)}$$

or

$$\left( \frac{u_c(y)}{y} \right)^2 = \sum \left( \frac{c_i u(x_i)}{y} \right)^2 = \sum \left( \frac{c_i x_i}{y} \frac{u(x_i)}{x_i} \right)^2 = \sum \left( c_i^{\text{rel}} \frac{u(x_i)}{x_i} \right)^2$$

## 2. UNCERTAINTY ESTIMATION PROCESS

### 2.1. Step 1 & 2 — specification & identify uncertainty sources

Most of radiochemical measurement results are obtained at the end of the procedure, which yields a numerical value for the measurand (the quantity subject to measurement) that is depend upon the number of intermediate or input quantities. These may be other measurands or constants (constants have also uncertainties).

The measurand has a relationship to these other quantities which, in principle can be expressed algebraically as  $y = f(\dots, x_i, \dots)$ . Such a formal approach can help in certain circumstances but in practice is more usual to break down the measurement procedure into simple blocks. The results of the uncertainty calculated in each block are used for calculating the combined uncertainty.

For uncertainty calculation it is advisable to break down the relationship between measurand and the input quantities.

### 2.2. Step 1- specification

Most of radiochemical measurement results are obtained at the end of the procedure, which yields a numerical value for the measurand that is depend upon the number of intermediate or input quantities. These may be other measurands or constants (constants have also uncertainties).

The measurand has a relationship to these other quantities which, in principle can be expressed algebraically as  $y = f(\dots, x_i, \dots)$ . Such a formal approach can help in certain circumstances but in practice is more usual to break down the measurement procedure into simple blocks. The results of the uncertainty calculated in each block are used for calculating the combined uncertainty.

For uncertainty calculation it is advisable to break down the relationship between measurand and the input quantities.

In the simple case of calculation of specific activity of  $^{90}\text{Sr}$  in an aliquot of spent resin performing separation of Sr by chromatographic method and measured by LSC.

$$A_e = \frac{N}{\text{Eff} \cdot \text{CY} \cdot M}$$

where

<b>A<sub>e</sub>:</b>	Specific activity of the nuclide in the sample
<b>N:</b>	Net count rate of sample
<b>Eff:</b>	Efficiency
<b>CY:</b>	Chemical Yield
<b>M:</b>	Mass/volume of original sample determined

But these terms are derived from other expression, which are related with other parameters; the efficiency, for instance, is calculated experimentally counting a reference sample (RS) with know activity ( $A_{RS}$ ).

$$\text{Eff} = \frac{N_{RS}}{A_{RS}} \cdot [1 - \text{EXP}(-\lambda_{Y-90} \cdot \Delta t)]$$

being

$$N_{RS} = R_{RS} - B$$

In previous equation we can find a term  $[1 - \exp(-\lambda_{Y-90} \cdot \Delta t)]$  to take into account (discount) the beta emissions of  $^{90}\text{Y}$  growing from  $^{90}\text{Sr}$  separation ( $\Delta t$ ):

where

- $N_{RS}$** : Net count rate of reference sample
- $A_{RS}$** : Activity of reference sample
- $R_{RS}$** : Count Rate of reference sample
- $B$** : Background (Blank) count rate
- $\lambda_{Y-90}$** : Disintegration constant of  $^{90}\text{Y}$
- $\Delta t$** : Lap time from separation to counting.

Chemical yield calculations are performed using the following expression that relates the mass of carrier added in the beginning of separation process (in the case that the previous determinations of total Sr content in the sample were negligible) and the same at the end:

$$\text{CY} = \frac{m_f}{m_i}$$

where,

- $m_i$** : Carrier mass added to the sub-aliquot
- $m_f$** : Carrier mass recovered in the measured sample

Finally the activity must be normalised to the equivalent mass of original sample determined in the sub-aliquot. To calculate this term we must take into account not only the mass of dry resin weighted ( $m_{alq}$ ) under prescribed conditions, but the different dilutions and concentrations carried out in taking the aliquot and sub-aliquot and in the measurement sample preparation.

$$M = \frac{m_{alq} \cdot DF_2}{DF_1}$$

- $m_{alq}$** : Mass/volume of the aliquot of original sample
- $DF_n$** : Dilution/concentration factors

Net count rate of sample is derived form the counting of prepared sample as following:

$$N = R - B$$

Being:

**R:** Gross count rate of the sample.

### 2.3. Step 3 & 4- quantify uncertainty components

In order to perform a good uncertainty assessment, it is useful to fulfil the uncertainty budget for each calculation process described. (See tables at the end of this publication).

We will start with the calculation of the uncertainty associated with the reference source counting due to it is a sum of terms we absolute combined square uncertainty is the summation of absolute variances of the terms involved:

$$U_N^2 = U_R^2 + U_B^2$$

The process of background measurement involved several (n) measurements of a blank sample during a time higher than the sample measurement time. So the uncertainty associated with this term can be calculated as experimental standard deviation:

$$U_B = \sqrt{\frac{\sum_{j=1}^n (B_{RS_j} - \bar{B})^2}{n-1}}$$

And the same for the gross count of the reference sample if more than two replicates of the counting process have to be carried out. On the other hand the deviation of Poisson distribution have to be applied:

$$U_R = \sqrt{\frac{\sum_{j=1}^n (R_{RS_j} - \bar{R})^2}{n-1}} \quad \text{or} \quad U_R = \sqrt{R_{RS}}$$

Due to the relation between R and B with N is a summation the absolute approach to obtain the sensitivity coefficients for combined uncertainty calculation is easier than the relative approach:

$$\begin{aligned} N &= R - B \\ c_R &= \frac{\partial N}{\partial R} = 1 \quad c_B = \frac{\partial N}{\partial B} = -1 \\ U_N^2 &= c_R^2 \cdot U_R^2 + c_B^2 \cdot U_B^2 = U_R^2 + U_B^2 \end{aligned}$$

Efficiency calculation, in fact depends on the  $N_{RS}$  and other parameters as the activity of the reference sample (certificate value) and the decay constant, taking into account the  $^{90}\text{Y}$  generation in the lap from the strontium separation to counting time. The uncertainty from the first parameter is type A due to we can evaluate by statistical methods and the others are type B uncertainties.

Due to the relationship between Eff and N, A<sub>RS</sub>, and (1-exp (-λt)) are a product the best approach to calculate the sensitivity coefficients for combined uncertainty calculation is the relative one.

In this case we can consider negligible the uncertainty coming from decay/growing factor of <sup>90</sup>Sr/<sup>90</sup>Y due to the uncertainties from the value of λ and the lap time are considered negligible also.

$$\begin{aligned} \text{Eff} &= \frac{N_{RS}}{A_{RS}} \cdot [\text{EXP}(-\lambda \cdot \Delta t)] \\ c_{N_{RS}} &= \frac{\partial \text{Eff}}{\partial N_{RS}} \cdot \frac{N_{RS}}{\text{Eff}} = \frac{[\text{EXP}(-\lambda \cdot \Delta t)]}{A_{RS}} \cdot \frac{N_{RS}}{\text{Eff}} = 1 \\ c_{A_{RS}} &= \frac{\partial \text{Eff}}{\partial A_{RS}} \cdot \frac{A_{RS}}{\text{Eff}} = \frac{-N_{RS} \cdot [\text{EXP}(-\lambda \cdot \Delta t)]}{A_{RS}^2} \cdot \frac{A_{RS}}{\text{Eff}} = -1 \\ \left[ \frac{U_{\text{Eff}}}{\text{Eff}} \right]^2 &= \left[ c_{N_{RS}} \cdot \frac{U_{N_{RS}}}{N_{RS}} \right]^2 + \left[ c_{A_{RS}} \cdot \frac{U_{A_{RS}}}{A_{RS}} \right]^2 \end{aligned}$$

Another interesting result is the one obtained for the calculation of relative mass of the original sample measured in which different to normal distributions of the uncertainties type B have to be combined. In this specific case the dilution factor for the mineralised sample and the volume taking for measurements are taking from the producer data of the beaker and the pipette respectively. These give semi-range values of a uniform (rectangular) probability distribution whose divisor is  $\sqrt{3}$ .

AT the end of the calculation taking into account the divisor in function of the probability distribution and the input data and the sensitivity coefficients of each uncertainty component in terms of results units we get the standard deviation of each input data in the final result.

#### 2.4. Step 5- calculating the combined uncertainty

As is shown in the hands-out the relative and absolute combined uncertainties have to be calculated in the uncertainty budget.

Final stage is to multiply the combined standard uncertainty by the chosen coverage factor in order to obtain an expanded uncertainty. The expanded uncertainty is required to provide an interval which may be expected to encompass a large fraction of the distribution of values which could be reasonably to attributed to the measurand.

## 2.5. Uncertainty budget

### 2.5.1. Absolute uncertainty budget

Each parameter  $x_i$  has an input uncertainty  $\Delta x_i$  which is defined in the table e.g., it may be a semi-range of rectangular distribution or 95% estimate resulting from a normal distribution with a  $k=2$  coverage factor etc.

Symbol	Value $\pm$	Where the value given describes	Probability distribution	Divisor (Div)	$U(x_i)$	$c_i^{abs}$	$u_i(y)$ $\pm$
$x_i$	$= \square x_i$ in $x_i$ units			To convert input $\square x_i$ to $u(x_i)$	In units of $x_i$ [ $= \square x_i / Div$ ]	Sens. Coeff for absolute values	In units of $y$ $= [c_i^{abs}$ $* U(x_i)]$

$\%U(x_i)$  means the absolute standard uncertainty.

### 2.5.2. Relative uncertainty budget

Each parameter  $x_i$  has an input uncertainty  $\Delta x_i$  which is defined in the table e.g., it may be a semi-range of rectangular distribution or 95% estimate resulting from a normal distribution with a  $k=2$  coverage factor etc.

Symbol	Value $\pm \%$	Where the % value given describes	Probability distribution	Divisor (Div)	$U(x_i)$ $\pm \%$	$c_i^{rel}$	$u_i(y)$ $\pm \%$
$x_i$	$= \Delta x_i / x_i * 100$			To convert input $\% \Delta x_i$ to $\% u(x_i)$	$= [U(x_i) /  x_i $ $]* 100$ $=$ $[\Delta x_i * 100 / (x_i$ $* Div)]$	Sens. Coeff for relative values	$= [U$ $(y) /  y  ] * 100$ $= [c_i^{rel}$ $* U(x_i) /  x_i  ] * 1$ $00$

$\%U(x_i)$  means a percentage of relative standard uncertainty, defined as  $U(x_i) / |x_i| * 100$ , where  $U(x_i)$  is the standard uncertainty.

N <sub>RS</sub> Input value uncertainty and distribution							
Symbol	Value ±	Where the value given describes	Probability distribution	Divisor (Div)	U(x <sub>i</sub> )	c <sub>i</sub> <sup>abs</sup>	u <sub>i</sub> (y) ±
x <sub>i</sub>	= Δx <sub>i</sub> in x <sub>i</sub> units			To convert input Δx <sub>i</sub> to u(x <sub>i</sub> )	In units of x <sub>i</sub> [=Δx <sub>i</sub> /Div]	Sens. Coeff for absolute values	In units of y =[ c <sub>i</sub> <sup>abs</sup> * U(x <sub>i</sub> )]
R <sub>RS</sub>	25.94	U type A	Normal	1	25.94	1	25.94
B	0.01	U type A	Normal	1	0.01	-1	-0.01
U <sub>N</sub>		Combined absolute uncertainty					25.94
U <sub>N(%)</sub>		Combined relative uncertainty					1.4 %

Eff Input value uncertainty and distribution							
Symbol	Value ± %	Where the % value given describes	Probability distribution	Divisor (Div)	U(x <sub>i</sub> ) ±%	c <sub>i</sub> <sup>rel</sup>	u <sub>i</sub> (y) ±%
x <sub>i</sub>	= Δx <sub>i</sub> /x <sub>i</sub> *100			To convert input % Δx <sub>i</sub> to % u(x <sub>i</sub> )	= [U(x <sub>i</sub> )/ x <sub>i</sub>  ]*100 = [Δx <sub>i</sub> *100/(x <sub>i</sub> *Div)]	Sens. Coeff for relative values	= [U(y)/ y ]*100 = [ c <sub>i</sub> <sup>rel</sup> * U(x <sub>i</sub> )/ x <sub>i</sub>  ]*100
N <sub>RS</sub>	1.4	Combined U	Normal	1	1.4	1	1.4
A <sub>RS</sub>	1.5	Overall U	Normal	1	1.5	-1	-1.5
U <sub>Eff</sub>		Combined relative uncertainty					2.1

M Input value uncertainty and distribution							
Symbol	Value ± %	Where the % value given describes	Probability distribution	Divisor (Div)	U(x <sub>i</sub> ) ±%	c <sub>i</sub> <sup>rel</sup>	U <sub>i</sub> (y) ±%
x <sub>i</sub>	= □ x <sub>i</sub> /x <sub>i</sub> *100			To convert input % □ x <sub>i</sub> to % u(x <sub>i</sub> )	= [U(x <sub>i</sub> )/ x <sub>i</sub>  ]*100 = [□ x <sub>i</sub> *100/(x <sub>i</sub> *Div)]	Sens. Coeff for relative values	= [U(y)/ y ]*100 = [ c <sub>i</sub> <sup>rel</sup> * U(x <sub>i</sub> )/ x <sub>i</sub>  ]*100
m <sub>ald</sub>	4.11E-3	Uncertainty	Normal	1	4.11E-3	1	4.11E-3
FD <sub>1</sub>	11	Semi-range	Rectangular	√3	6.4	-1	-6.4
FD <sub>1</sub>	2.0	Semi-range	Rectangular	√3	1.2	1	1.2
U <sub>M</sub>		Combined relative uncertainty					6.5

**CY** **Input value uncertainty and distribution**

Symbol	Value ± %	Where the % value given describes	Probability distribution	Divisor (Div)	U(x <sub>i</sub> ) ±%	c <sub>i</sub> <sup>rel</sup>	U <sub>i</sub> (y) ±%
	= □ x <sub>i</sub> /x <sub>i</sub> *100						
m <sub>f</sub>	1.2	Uncertainty	Normal	1	1.2	1	1.2
m <sub>i</sub>	1.0	Uncertainty	Normal	1	1.0	-1	-1.0
U <sub>CY</sub>	Combined relative uncertainty						

**N** **Input value uncertainty and distribution**

Symbol	Value ±	Where the value given describes	Probability distribution	Divisor (Div)	U(x <sub>i</sub> ) In units of x <sub>i</sub> [= □ x <sub>i</sub> /Div]	c <sub>i</sub> <sup>abs</sup>	u <sub>i</sub> (y) ±
	= □ x <sub>i</sub> in x <sub>i</sub> units						
R	1.64	U type A	Normal	1	1.64	1	1.64
B	0.01	U type A	Normal	1	0.01	-1	-0.01
U <sub>N</sub>	Combined absolute uncertainty						
U <sub>N</sub> (%)	Combined relative uncertainty						

**Asr** **Input value uncertainty and distribution**

Symbol	Value ± %	Where the % value given describes	Probability distribution	Divisor (Div)	U(x <sub>i</sub> ) ±%	c <sub>i</sub> <sup>rel</sup>	U <sub>i</sub> (y) ±%
	= □ x <sub>i</sub> /x <sub>i</sub> *100						
N	1.2	U type A	Normal	1	4.0	1	4.0
Eff	1.0	Comb. Uncertainty	Normal	1	2.1	-1	-2.1
CY	1.0	Comb. Uncertainty	Normal	1	1.6	-1	-1.6
M	1.0	Comb. Uncertainty	Normal	1	6.5	-1	-6.5
U <sub>Asr</sub>	Combined relative uncertainty						
U <sub>Asr</sub>	Expanded relative uncertainty (ej. k = 2)						



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