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Lifetime measurements of ionic electroactive polymer actuators

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Abstract
This article is focused on proposing a unified methodology for automating the measurement procedures of ionic electroactive polymer actuators. The proposed methodology and large-scale automation would make testing ionic electroactive polymer actuators less labor-intensive and allow analyzing many ionic electroactive polymer actuators simultaneously. Defining a clear framework for testing ionic electroactive polymer actuators performance and reliability would make the testing process reproducible and provide better comparison between ionic electroactive polymer actuators of either different or similar classes. Our methodology separates two types of degradation: degradation during operation and spontaneous self-degradation.

Keywords
Actuator, electroactive polymers, structural health monitoring

Motivation

Current technologies where moving parts are achieved through electromechanical, pneumatic, or hydraulic actuators often make products too noisy, heavy, and complicated. Many fields show a notable demand for soft, simple, and miniature actuation devices. The ionic electroactive polymer actuators (IEAPs) are good candidates for this purpose. Although research has continued for a few decades, challenges hindering their commercial success remain.

The layered bending IEAPs provide a large bending deformation even at low (~1%) intrinsic strains of the electroactive .layer, at low driving voltages (0.1–5 V) (Bahramzadeh and Shahinpoor, 2013; Jo et al., 2013; Kosidlo et al., 2013; Temmer et al., 2013). A major goal in soft polymeric electromechanical actuator research is to utilize their lightness: low driving voltage, ease of miniaturization, high power-to-weight ratio, work density, and silent operation. Furthermore, the softness of IEAPs and tunable properties would allow for a variety of operation modes, which could redefine what kind of moving and controllable parts can be manufactured.

In addition to electromechanical or electro-chemomechanical bending, an IEAP actuator can simultaneously sense motion (Pugal et al., 2010) or humidity (Must et al., 2013), harvest energy (Tiwari and Kim, 2013), and even store electric energy as a supercapacitor (Torop et al., 2011). To utilize IEAPs' low weight, the best application areas would be satellites and space crafts (Krishen, 2009), medicine, and lab-on-chip devices (Carpi and Smela, 2009). A variety of IEAP-based applications have been proposed during the recent decade (Carpi et al., 2011; Kim and Tadokoro, 2007). As a rule, all of them have demonstrated their capabilities for a limited time, working close to their theoretical performance limit. However, besides the actual performance of the IEAP devices, their acceptance in critical applications is currently limited by the lack of research in their reliability, durability, and reproducibility.

Reliability has been recognized as the most critical attribute limiting the acceptance of IEAPs in space systems and medical systems—fields that could benefit most from their lightness. Any malfunction in space technology would be a tremendous financial loss and in case of a habited station or craft danger the people on the mission. Malfunction of a medical device such as a prosthesis is serious risk for the patient. In both cases,

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worst-case scenario may lead to human casualties. For IEAPs to become accepted in the fields that have most to gain from them, a reliable testing framework for individual pieces should be established.

To begin establishing clear rules by which to analyze performance numbers, a large statistical sample size is required. However, the state-of-the-art IEAPs have not reached mass production or commercial applications yet. Usually, the results published in scientific articles and reports are obtained from a few individual samples only. The samples are manufactured in laboratory conditions as small batches. The fabrication resembles cooking, requiring expertise, experience, and skillful hands. The developed devices are often irreproducible or use technologies and materials, which are expensive and/or not viable for industrial production. Although few researchers dare to admit that production is irregular (Liu et al., 2007), it is obvious that even no two samples from the same laboratory are identical. As a result, the situation with the IEAPs is very different from that, say, the confectionery industry, where millions of identical biscuits are always available for testing.

In our laboratory, where we are fine tuning IEAP fabrication methods, the quantity of samples is difficult to handle. In order to compare the individual samples on similar terms and perform lifetime tests, we developed an automatized large-scale testing equipment to analyze many, up to hundreds of actuators simultaneously. This setup minimizes human error and guarantees that all samples are tested in comparable conditions. Moreover, this setup is able to compare different IEAP types using a unified methodology.

During the last two decades, the assortment of IEAPs has evolved from the aqueous ionic polymer–metal composite (IPMC) and "wet" conducting polymer (CP) actuators to a rich variety. Thousands of articles about the various IEAPs have been published in the recent decade reporting advancements such as better strain and speed, or smaller dimensions. However, only a few of them take into account the lifetime, stability, or cycle life of their samples, and the reported data are seldom comparable. It is commonly recognized that lifetime or cycle life of IEAPs is highly dependent on strain and stress, while frequency dependence relates to conductivity or diffusion speed (Bennett, 2004). At large, the reported cycle life of different IEAPs varies between thousands and millions of cycles. Some authors praise the large strain of their samples, while cycle life tests are implemented at high frequency and almost imperceptible strain. Obviously, the reason behind data discrepancy is the absence of a generally recognized methodology.

The cycle life has been most thoroughly studied on CP-type IEAPs. Vidal et al. (2006) report that their actuators standing of interpenetrating polymer network actuators containing an electronic CP can be cycled for 10^4 times at 1 Hz and 3.5×10^6 times at 10 Hz. Lu et al. (2002) just stop the lifetime test of the CP actuator devices after 10^4 cycles. Madden et al. (2000, 2007) have performed thorough research and argue that the cycle life of polypyrrole actuators depends on strain, reaching 1.2×10^5 cycles with very low strain at 3 Hz and 10^3 cycles at high strain and 32×10^3 at low strain at the frequencies of 0.2–1 Hz. Liu et al. (2007) describe the performance loss of similar actuators as 20% loss after 10^4 cycles, 50% at 3.5×10^4, and virtually no electro-chemo-mechanical activity by 6×10^4 cycles. Contrarily, Bennett and Leo who work with Nafion–platinum IPMC compare water and ionic liquid as electrolyte and report the cycle life being 10^3 for the former and 3×10^5 for the latter (Bennett, 2004). According to Du et al. (2011), their ionic polymer–carbon nanotube composite actuators can last for 3000 cycles with only 10% reduction in the displacement output.

The purpose of this article is sharing our experiences and best practices emerged in the course of designing and the use of automatic IEAP testing equipment. As the design and capabilities are based on a trade-off between automation level, ease of use, and the quality acquired data, the focus of this article is justifying the design and choice of testing parameters to best characterize IEAPs. In terms of long-term degradation, the evolutionary trends common to all IEAP types rather than the comparative data analysis or formal statistics of all particular samples are given. Our testing methodology describes IEAP degradation well and we recommend it as standard procedure.

Method

Pre-requisites

The core in IEAP actuators is a pliable porous polymer film, imbued with an appropriate liquid electrolyte. Obviously, a structure like this will not last forever, due to the decomposition of the polymer backbone and evaporation or leakage of electrolytes. As a result, the performance of IEAPs decreases in time. The rate of performance drop depends mainly on the type of work that has been done, such as the number of performed work cycles; working frequency and recurrence; and applied load, strain, and stress, and also on environmental conditions—temperature, humidity, and so on. It is likely that some of the factors remain unknown.

Long-term experiments with IEAPs show two types of degradation:

- *Degradation during operation*—fatigue of membrane and electrodes, as well as leakage of electrolyte out of membrane caused by mechanical deformation.
- *Spontaneous self-degradation*—long-term alteration of parameters caused by the ambient environment. We noticed spontaneous self-

degradation even when samples were held in conditions suggested by the manufacturer, for example, immersed in an electrolyte.

General test procedure

IEAPs generally have a long lifetime expectancy (sometimes referred to as "cycle life") of 10^4–10^7 working cycles. We are interested in how their performance changes after use. It is not necessary to measure each cycle. It is enough to test each sample after a while of working. Therefore, optimal testing should separate training (that would simulate continuous use) and measurement as two separate procedures.

During training, an actuator is excited with the appropriate voltage input, but its electrical and mechanical responses are not recorded. During measurement, the actuator is engaged with a wide frequency range signal, while its mechanical and electrical parameters of interest are recorded. The high cost of components in measurement station (camera, displacement sensor, force gauge, current booster, data acquisition (DAQ) system, etc.) justifies separating equipment for a more expensive test station and a less expensive training station. As a downside, moving between two stations makes the procedure more labor-intensive.

Testing a single actuator manually consists of the following steps:

1. The actuator is attached to electric contacts. (As samples are fragile, they should be handled carefully.) If a sample is reattached to contacts several times, the operator must ensure that all samples are positioned in the same orientation, and that the length of the sample between contacts stays constant.
2. The camera or laser displacement sensor, recording the bending response of the actuator, is focused to the sample. The actuator is excited with a pre-defined test signal. Simultaneously, the mechanical output, input voltage, and input current of the actuator are recorded.
3. The camera is replaced by a force sensor. The blocking force is measured only in the straight position. As in the previous step, the actuator is again excited with a similar test signal, and output signals are recorded.
4. The actuator is detached from the measurement station and attached to the separate training station, where all actuators are excited with similar sine voltage of constant frequency. Special care is paid to accounting the exact number of passed working cycles on each particular sample.

This testing process is repeated until decided to terminate, based on the extent of degradation.

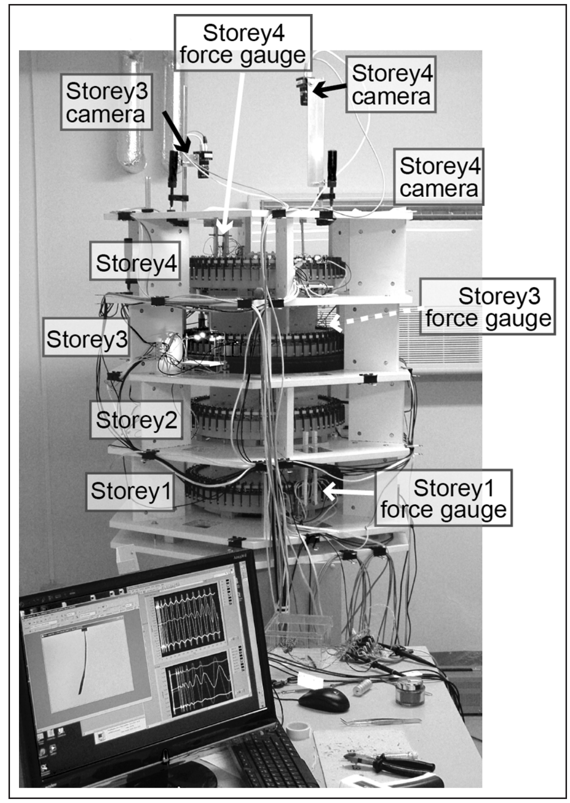

Figure 1. The four-level conveyor.

Large-scale test equipment

Working principle

The described testing procedure requires that an operator acts with persistent precision. One operator is able to test only 10–20 samples simultaneously, performing about 10 cycles on each sample in a day. Therefore, we designed a testing equipment to perform the described testing process automatically and work on many actuators simultaneously. Our setup excludes human error and guarantees that all samples are tested in exactly similar conditions. Its capabilities are based on a trade-off between robustness and the amount and type of acquired data, balancing it with an optimal level of automation and ease of use.

The test bench is seen on Figures 1 and 2. It is a circular conveyor of four levels, sharing the same rotary axis and acting synchronously. Each level can hold up to 60 actuators of similar type. Each level can perform a specific testing sequence, so the whole setup can test up to 240 samples of up to four different types. The actuators are attached to clamps, hanging around the turntable during measurement processes. This method avoids their damage and ensures that the samples are not turned over between the cycles.

Our equipment is located in a separate room equipped with an air conditioner of sufficient capacity to guarantee the stable temperature (17°C) and humidity level (30% relative humidity (RH)) of the ambient air for the duration of the whole experiment.

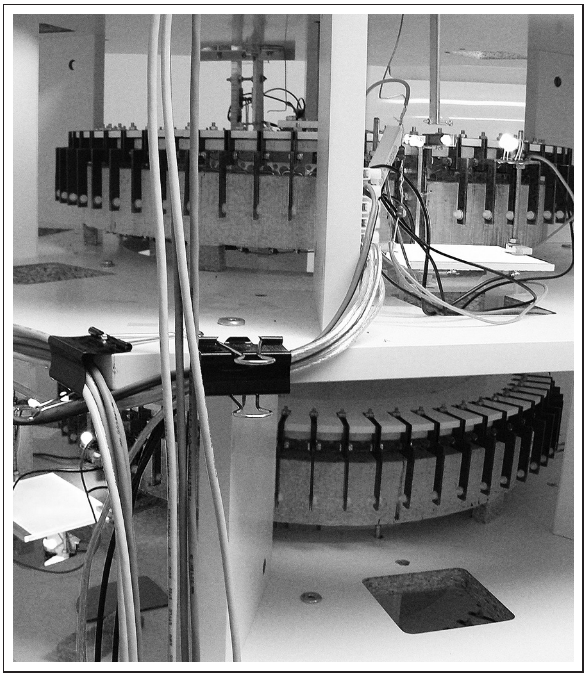

Figure 2. Two levels of the test bench with hanging clamps.

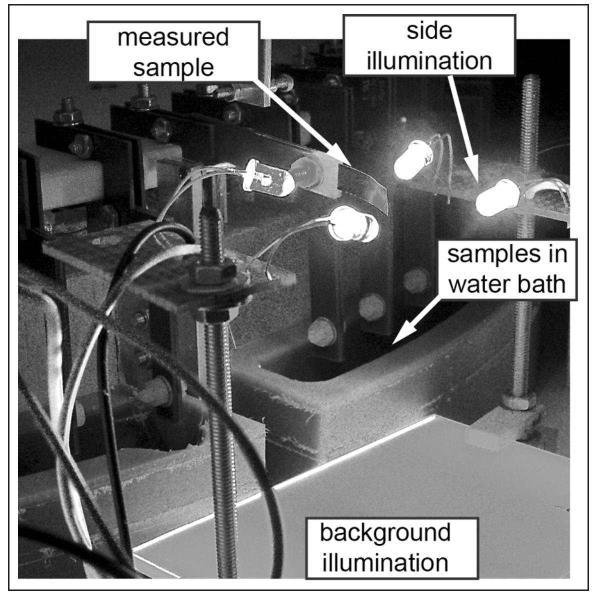

Figure 3. For measurement, a sample is lifted out of the water bath. The proper illumination is essential.

Four samples are concurrently measured by cameras (one for each level), and four samples are measured with force sensors (one for each level). After performing a measurement cycle, the circular conveyor turns for 6°, sets the eight samples, just tested, for training, and positions the next eight samples for testing.

Measurements are carried out in cantilever configuration—one end of an actuator is fixed, while rest is free to move or to apply force to the force gauge. During measurement cycles, samples are held horizontally and edgewise—so distortion from the actuator's self-weight has less effect on measurements. During training cycles, samples are held hanging vertically. A vertical position is necessary to negate creep—permanent deformation under the influence of inner stresses, which is experienced by some types of IEAPs. When an actuator is hanging freely, its own weight diminishes this effect. Second, some types of IEAPs must be continuously in wet environment, others have to be moistened in water or some solvent after every certain time. Therefore, vertical placement of samples allows soaking the wet types of IEAPs in their solvent bath. To avoid drying wet actuators, the measurement cycle that is carried out in ambient environment should not exceed a few minutes, covering all required frequencies. An example of an actuator, lifted out of wet environment, is depicted in Figure 3.

Technical details

Only the samples under testing are positioned horizontally, while all remaining samples under training are hanging vertically. The process of turning samples between two positions is explained in Figure 4. Initially, a sample is hanging vertically, possibly in a container of solvent, while the turntable squeezes the electrical terminals of the clamps against properly positioned electric contacts (Figure 4(a)). In order to align the sample horizontally, the turntable and its clamps lift up and turn, positioning a particular clamp over a tubular support—"the lower support." While lowering, a turntable turns the clamp up (Figure 4(b)), until electrical terminals are squeezed against appropriate spring contacts (Figure 4(c)). Now, the sample is ready for testing—in the camera's field of view or pushed against a force sensor. The state of remaining 58 samples is as in Figure 4(a). Aligning a sample back vertically is performed by lifting the turntable up and pushing the clamp against the upper tubular support. Next, the turntable is rotated by 6°—lifting it down arranges the just measured sample in the alignment depicted in Figure 4(a) and sets the next one under measurement (Figure 4(c)). This mechanism allows eight samples (two on each of the four levels) to be switched between two alignments synchronously—a rotating turntable enables measuring all samples, one after another.

Special attention was paid to contact clamps: avoiding contamination of the electroactive materials. The contacts should be made of noble metals (gold or platinum). Initially, clamps were fabricated as printed circuit boards. The gilded electrode surfaces were obtained by standard surface plating used for printed circuit boards—electroless nickel immersion gold (ENIG) method. However, according to the manufacturer's data, the layer of gold, which has been plated by ENIG method, is only 0.05–0.125 μm thick, underneath is a 3- to 6-μm nickel layer on a 35-μm copper layer. As

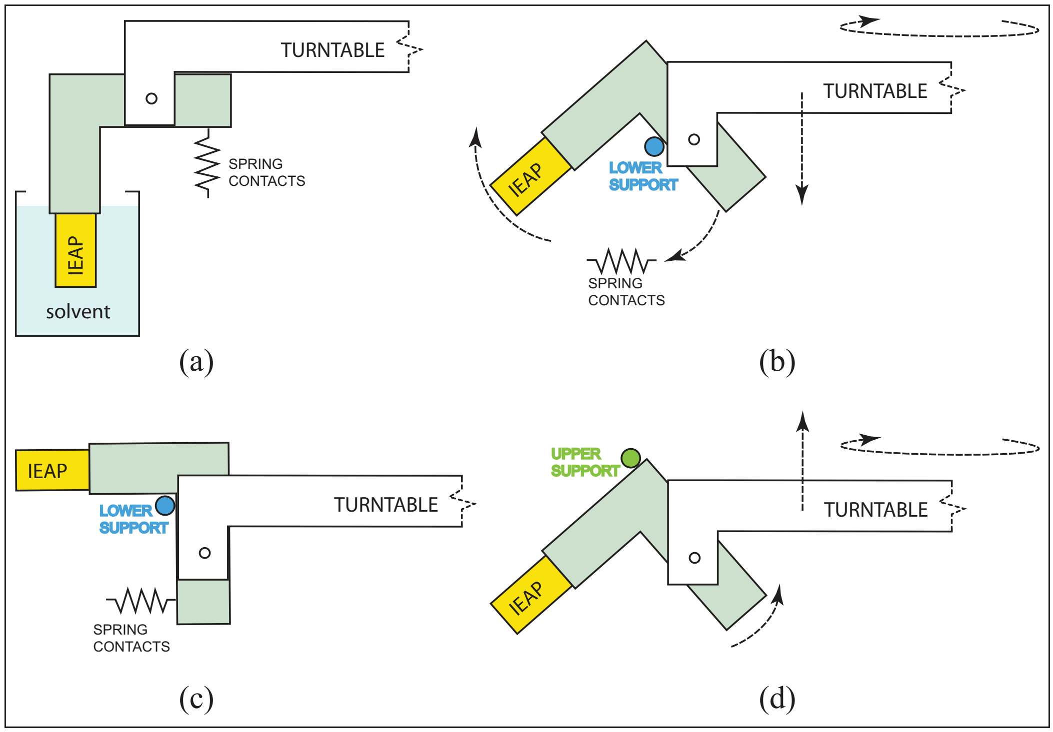

Figure 4. Repositioning the samples between vertical and horizontal alignments: (a) training cycle (actuator is positioned vertically), (b) lifting sample up, (c) measurement cycle (actuator is positioned edgewise and horizontally), and (d) lifting sample down.
IEAP: ionic electroactive polymer actuator.

shown in Figure 5(a), even a gentle touch can rub off the thin gold and reveal the electrochemically active nickel or even copper. So, the contact clamps were reworked, soldering to clamps a 1.5-mm-wide ribbon of 50-µm-thick sheet of gold. The solder and opposite surface were covered with a resistant epoxy lacquer. The *best practice* contact clamps are depicted in Figure 5(b).

Recorded mechanical output

The mechanical output of an IEAP's blocking force and its characteristic bending have to be recorded. The most reliable way of describing the transient behavior of IEAPs is recording bending with a camera and processing this visual information. The bending behavior of the actuators was recorded by cameras, specifically monochrome USB cameras (DMK 22BUC03) equipped with a lens of long focal length to reduce any parallax effect. Properly calibrated lighting on samples from back and two sides as depicted in Figure 3 simplifies image processing.

Measuring blocking force may seem like a simple and cheap task as the output from the force sensor is a single voltage. However, automation of this approach is applicable with fully straight actuators only. Automated manipulation on crooked actuators has two drawbacks: (a) its automated positioning to close

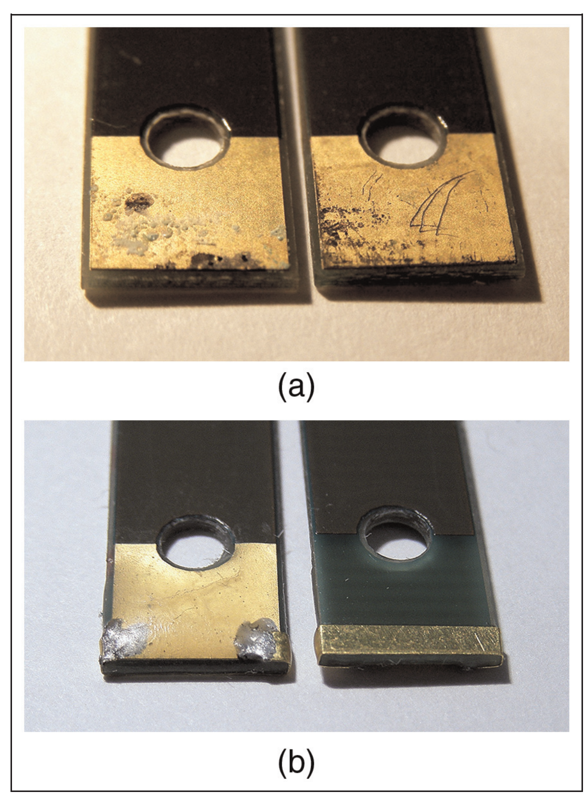

Figure 5. Contact clamps: (a) unsuitable and (b) *best practice*. The width of the contact clamp is 8 mm.

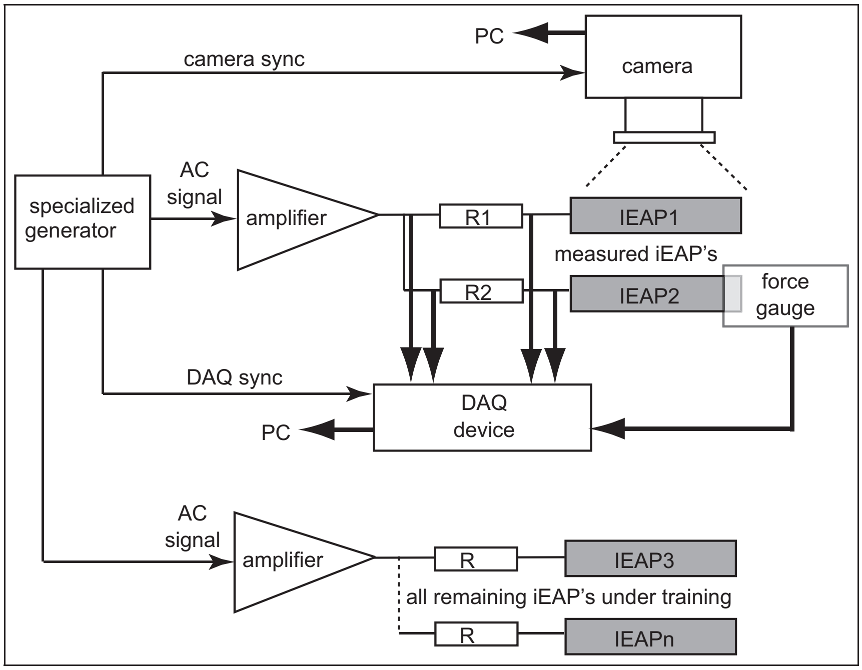

Figure 6. Schematics of a single level on the conveyor.
AC: alternating current; DAQ: data acquisition; IEAP: ionic electroactive polymer actuator.

proximity of a force gauge may easily damage the sample and (b) it rarely gets exactly in the middle of the measuring range.

Schematic of the measurement

The schematic of the experiment setup is given in Figure 6. Similar configurations are described in our previous works (Punning et al., 2009; Vunder et al. 2012). In order to reduce load on computer, voltages are generated by a specialized generator using an Atmel microprocessor. The generator produces an alternating current (AC) signal for driving actuators being tested and triggers for synchronizing camera and the DAQ device. Using this special generator allows synchronizing measurements and camera with the input signal at minimal sampling rate for each particular frequency. Input signals are amplified by custom-made current amplifiers, based on the Burr-Brown OPA548 operational amplifier.

In conclusion, the four-storey conveyor incorporates the following:

- *Five special generators*—one for each level and one for generating the common training cycles.
- *Eight current amplifiers*—two for each levels, one for measurements and one for training.
- *Four USB cameras*—one for each level (DMK 22BUC03 of The Imaging Source).
- *Four force gauges*—one for each level (Millinewton of EPFL or MLT0202 of ADInstruments).
- *One motorized linear actuator*—to lift the circular turntable up and down within 5 cm (Thorlabs TravelMax Stage LNR50VK1/M).
- *One motorized rotational stage*—to rotate the turntables continuously with respect to the vertical axis (Thorlabs CR1/M-Z7E).

The resistors "R" at each IEAP under training protect the amplifier from occasional short circuit on actuators being tested. All electrical signals—voltages, electric currents, and outputs of force gauges—are registered by a single DAQ card with 32 analog inputs (National Instruments PCI-6254). The electric current output is measured as a voltage drop on a low-ohm resistor, chosen according to the IEAP type. The whole system is operated by a single PC-type computer and National Instruments LabVIEW software.

Driving signal

The experiment setup as described above generates a huge amount of data that are difficult to handle and complex to process. Therefore, special attention was paid to the driving signal—a waveform of voltage that is applied to the actuators for testing. It was designed with several careful considerations, which are explained as follows.

Measuring "wet" IEAPs in dry environment should be performed as fast as possible. ("Dry" EAPs have no such restriction.) Conventional aqueous IPMC can work in air for only a few minutes before its parameters

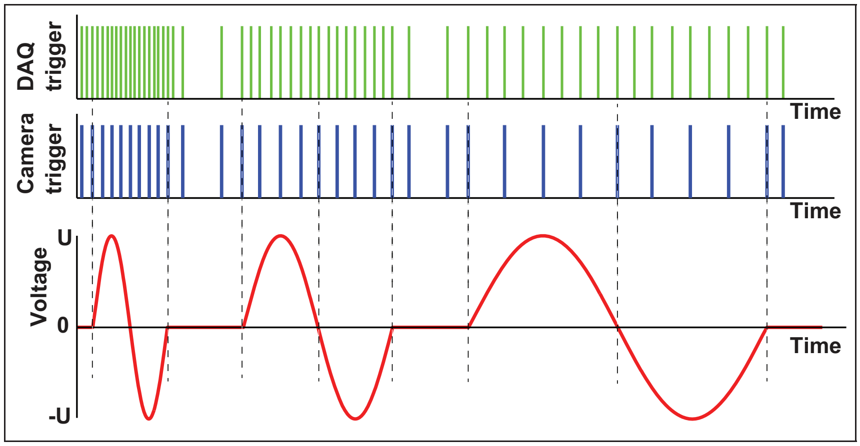

Figure 7. Gradual sweep waveform and triggering pulses.
DAQ: data acquisition.

change drastically due to drying. Therefore, a driving signal should be as short as possible.

To choose an appropriate driving signal frequency, we first remarked that IEAP actuators are capable of working at a limited range of frequencies from direct current (DC) to a few tens of hertz. To recognize the frequency dependence of degradation, the bandwidth of the driving signal should meet the frequency range of the actuators. For electrical measurements, commonly sweep frequency sine or white noise is used at a high sampling rate, chosen according to the Nyquist–Shannon sampling theorem.

An experiment setup described above generates an enormous amount of data. The volume depends on the DAQ rate, which is commonly chosen based on the highest measured frequency and taken as a basis for the entire experiment. For electrical measurements, the number of measurement samples makes no problem as the data are small in comparison to the video feed from cameras. To receive all frames from a camera without any fault, they must be temporarily stored in the computer's random access memory (RAM). The amount of RAM is a limiting factor to the total number of video frames recorded during a single measurement cycle. To limit the size of video data without setting constraints to the frequency range, the driving waveform was a gradual sweep. This approach allows capturing video and performing electrical measurements with (varying) dynamic frame rates that adapt to the particular frequency of the analog driving signal. The synchronization between DAQ and camera triggering with the driving waveform was accomplished using a custom-made signal generator. An illustrative example of the gradual sweep with the corresponding triggering pulses is presented in Figure 7.

In our setup, the driving voltage waveform is a gradual sweep of sine signals, lasting approximately 2.5 min. The frequencies are (in this order) 10, 5, 2.5, 1.25, 0.64, 0.32, 0.16, 0.08, and 0.04 Hz. Each frequency lasts for three half-periods, while the initial phases are opposite. The purpose of the 0.5 half-period delays between the sine waveforms is the interim discharging of capacitive actuators. Using the setup described, the number of frames, captured by the camera, is reduced to 180, and the number of DAQ samples is reduced to 840, while the actuator performs only 13 bidirectional bending cycles.

Reduction in the video data

As an uncompressed video feed from each measurement cycle is considerably large, the required disk space can easily grow into terabytes. Therefore, it is rational to process the video data immediately, discard the raw data, and leave only a selection of numerical parameters, necessary to restore the shape of actuator with sufficient accuracy. A convenient method is the vector interpretation, described in our previous works (Punning et al., 2009; Vunder et al. 2012).

Vector interpretation of the actuator's shape expresses its bending from input contacts along the sample. A curved line, representing the shape of the actuator, is divided into vectors of equal lengths, as depicted in Figure 8. Every vector represents the curvature at vector's origin. The shape of actuator is characterized by a number of angles $\theta 1$, $\theta 2$, …, θn, while each consecutive angle is presented with respect to the previous one. It is obvious that more vectors represent a complicated shape more accurately. However, the accuracy of detecting the angles at each point and the general shape also depends on the resolution of the recorded image. In our experiment, the resolution of the camera feed was 640 $\times$ 480 pixels and the actuators were divided into nine vectors. Discarding images from camera and converting them into numeric data reduces the amount of required storage space significantly, as the data describing a single measurement are condensed to a few dozens of kilobytes only. Moreover, numeric data can be analyzed and compared better than visual images.

The image of an actuator under testing is recorded by a USB camera and processed by the appropriate National Instruments LabVIEW tool. A proper lighting and exposure as shown in Figure 3 greatly simplifies image processing and determining vectors. When camera is oriented transverse to the actuator and the stage is illuminated from the background, the image of the actuator becomes a single-contrast curved line. The side illumination, as shown in Figure 3, further helps to obtain a clear-cut image.

Performance of actuators can be determined from its bending characteristics. Most commonly used parameters are tip displacement, curvature, strain difference, and so on. A comprehensive overview about commonly used quantitative representations of the IEAP actuators is given by Kruusamäe et al. (2014). Vector interpretation, as described above, allows estimating any of them with high accuracy as well as developing qualitatively new parameters, according to the requirements and results of particular experiments.

Long-term degradation of the IEAPs

As mentioned, two types of IEAP degradation are recognized: degradation during operation and spontaneous self-degradation. Tests for long-term degradation were carried out in the following order:

Step 1. Initial conditions for each particular sample were recorded.
Step 2. After 10 weeks, the states of samples were recorded again.
Step 3. After 10 weeks from Step 2, degradation of samples from continuous loading was determined. The process was terminated when performance of most of the samples fell below a pre-determined value, commonly 5% of mean initial values.
Step 4. After about 2 weeks from Step 3, a final condition from samples was recorded again.

Between steps, the samples were kept in prescribed environmental conditions—each one in a separate vial and in an appropriate solvent if required.

We carried out a long-term degradation experiment on several types of IEAPs with identical methodology. The waveform of the driving signal, timing, and even dimensions (5×20 mm^2, except varying thickness) of all samples were constant. The only inconstant parameter was the excitation voltage's amplitude, which varied between IEAP types and was chosen according to the recommendations of the manufacturer. So, the tests at Step 3 took 3–30 days or 10^4–10^5 working cycles.

The subsequent analysis of the obtained data revealed that degradation in any type IEAP can be

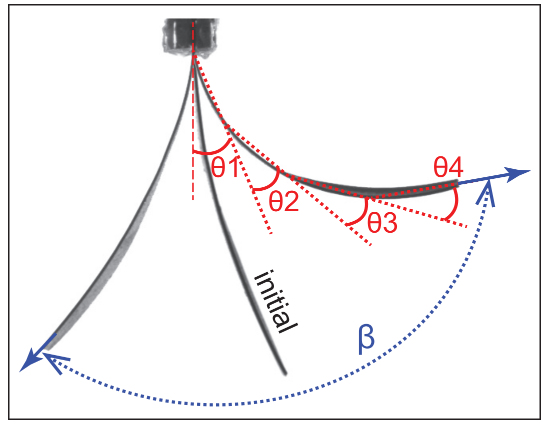

Figure 8. Shape of the actuator is described by angles $\theta 1$, $\theta 2$, …, θn and performance β.

conveniently evaluated from a single parameter—the peak-to-peak tip angle. It is the angle between the tip tangents measured from maximal bending displacements in opposite directions. This parameter, denoted as β in Figure 8, suits well for all IEAP types tested and will not fail even in the case of considerable creep. The obtained peak-to-peak tip angle can be plotted with respect to time as well as with respect to the performed work cycles. The former emphasizes the spontaneous self-degradation, while the latter shows degradation during operation.

As a detailed degradation analysis of IEAPs is beyond the scope of this article, we present a summary from our analysis of about 500 actuators from seven significantly different IEAP types. We conclude that IEAP actuators typically degrade in a predictable manner, as presented in Figure 9. The vertical axis β/β_0 denotes the peak-to-peak tip angle, normalized with respect to the first measurement (Step 1).

The left graph—time-dependent behavior—shows that IEAPs' performance decreases even when it is kept in idle state. Degradation continues at similar rate even after significant wear. The right graph of the Figure 9 shows that in the course of continuous working (Step 3), the performance decreases nearly exponentially with respect to the performed working cycles.

Conclusion

We have developed a methodology for testing durability of IEAPs, taking into account the two types of degradation—degradation during operation and spontaneous self-degradation. In our opinion, the described methodology is a good candidate for standard procedure of testing the durability of the IEAP actuators. The experiments carried out by this methodology and the large-scale equipment show that both types of degradation have serious effect on the IEAPs. Here comes the justification of the term "lifetime" of IEAPs over the term cycle life.

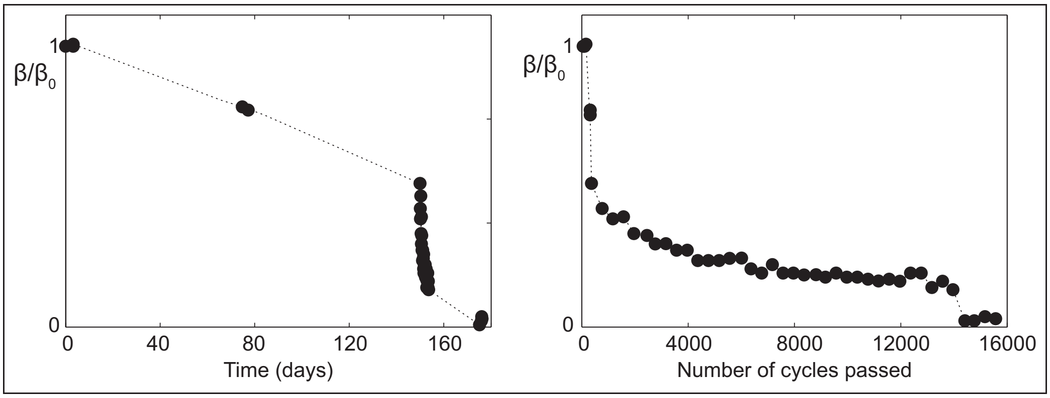

Figure 9. Typical pair of degradation charts of the IEAP actuators. Each point in the graphs marks one measurement cycle. To each point in the left graph corresponds one point in the right graph and vice versa.
IEAP: ionic electroactive polymer actuator.

Declaration of conflicting interests

The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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