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# Glass transition related cohesion of amorphous sugar powders

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## Abstract

The time-dependent nature of glass transition related cohesion was studied for amorphous sucrose, maltose, glucose, galactose and fructose powders. The powders were made by freeze drying and the low molecular weight sugars (glucose, galactose and fructose) were made as mixtures with lactose. The blow test was used to measure the cohesiveness of the different powders with respect to time while the powders were exposed to a constant temperature and relative humidity environment. It was found that the rate of change of cohesiveness of these powders was directly related to the amount that the glass transition temperature ($T_g$) was exceeded by ($T - T_g$). That is, cohesion was related to the combined effect of temperature and moisture and not the individual temperature, water activity or moisture content of the powder. This work confirms that the mechanism for sticking and caking of amorphous sugars is through the phase change of the amorphous sugar from a glass to a rubber at temperatures above the glass transition temperature. Furthermore, it confirms that the rate of cohesiveness development is proportional to the $T - T_g$ value, that is, the greater the temperature above the $T_g$, the quicker the powders will develop liquid bridges which may result in caking.

*Keywords:* Glass transition temperature; Stickiness; Caking; Amorphous sugar; Food powders

## 1. Introduction

The glass transition related changes of food powders containing amorphous sugars is well recognised. It has been attributed to causing stickiness and caking problems in a number of food powders including fruit powders (Bhandari, Datta, & Howes, 1997; Bhandari, Senoussi, Dumoulin, & Lebert, 1993; Brennan, Herrera, & Jowitt, 1971; Lazar, Brown, Smith, Wong, & Lindquist, 1956), sugar mixtures (Downton, Flores-Luna, & King, 1982; Wallack & King, 1988), coffee powder (Wallack & King, 1988) and dairy powders (Bronlund, 1997; Chuy & Labuza, 1994; Hennigs, Kockel, & Langrish, 2001; Lloyd, Chen, & Hargreaves, 1996; Özkan, Walisinghe, & Chen, 2002; Paterson, Bronlund, & Brooks, 2001; Paterson, Brooks, Bronlund, & Foster, 2005; Rennie, Chen, Hargreaves, & Mackareth, 1999; Roos & Karel, 1991a).

Food powders containing amorphous sugars experience stickiness and caking problems when the powder is exposed to temperatures above the powder's glass transition temperature ($T_g$), which is a function of the moisture content/water activity of the powder (Lloyd et al., 1996; Roos & Karel, 1991a, 1991b, 1991c, 1991d; Slade & Levine, 1991). At the $T_g$, the viscosity of the amorphous material decreases considerably allowing greater molecular mobility which results in sticky behaviour (Bhandari & Howes, 1999; Downton et al., 1982; Lloyd et al., 1996; Peleg, 1983; Roos & Karel, 1991a; Slade & Levine, 1991; Wallack & King, 1988). In general, stickiness, which is considered to be the onset of caking, occurs when liquid bridges of amorphous sugar, fatty or dissolved material composition form between adjacent powder particles (cohesion) or between the particle and another surface such as a spray dryer wall (adhesion) (Aguilera, del Valle, & Karel, 1995; Papadakis & Bahu, 1992; Peleg, 1983; Roos,

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1995). More advanced stages of caking occur as a result of the crystallisation of the liquid bridges (Aguilera et al., 1995; Roos, 1995). In this work, cohesion is used to describe sticking and caking of powder particles as part of a continual process, a process where it is difficult to separate the two from each other.

Most research on stickiness of amorphous powders consider sticking to occur at one point, i.e., for a given moisture content or water activity, a particular temperature will be required for stickiness to occur (Downton et al., 1982; Hennigs et al., 2001; Ozmen & Langrish, 2002). This temperature is usually referred to as the sticky point temperature. In this type of work, short time frames are used and often the temperature of the powder is increased quickly until a change in the cohesiveness of the powder is detected. The level of cohesion corresponding to "sticking" will be different for each method employed, depending on the sensitivity of each method and the manner in which the experiment was carried out, e.g., the rate that the water bath temperature is increased when performing the Lazar et al. (1956) test for stickiness. This is discussed further in detail by Paterson et al. (2001) and Paterson et al. (2005). This type of work is useful for measuring the temperature and RH conditions for instantaneous sticking as this information is valuable when dealing with sticking problems during spray drying. However, this work is not useful when considering sticking and caking problems during longer time periods, as is the case during storage.

Some researchers have proposed, and in some cases shown, that glass transition related flow changes (sticking, crystallisation and collapse) occur at a rate determined by the $T - T_g$, i.e., the amount that the $T_g$ has been exceeded by (Brooks, 2000; Levine & Slade, 1986; Lloyd et al., 1996; Paterson et al., 2001; Paterson et al., 2005; Roos & Karel, 1990, 1991a, 1992, 1993). Others have proposed that these glass transition related flow changes occur at a particular $T - T_g$ (Aguilera, Levi, & Karel, 1993; Hennigs et al., 2001; Ozmen & Langrish, 2002; Roos, 2002) which is true when this is in reference to instantaneous sticking, crystallisation or collapse. Brooks (2000), Paterson et al. (2001) and Paterson et al. (2005) investigated the rate of sticking of amorphous lactose by using different temperature and relative humidity (RH) conditions to give the same and different $T - T_g$ values. It was found that the rate of stickiness development was only dependent on the $T - T_g$. Two sets of temperature and RH conditions, which yield the same $T - T_g$, will result in the same rate of sticking. It was also demonstrated that the same level of cohesiveness could be achieved for a powder by using two different $T - T_g$ values if an appropriate amount of time was given in each case for the powder to reach the desired level of cohesiveness.

This work investigates the rate of change in cohesiveness, mainly stickiness, for other amorphous sugar powders (sucrose, maltose, glucose, galactose and fructose) in order to determine whether flowability problems associated with these sugars are related only to the $T - T_g$ and not the conditions used to achieve a particular $T - T_g$. If cohesion is only related to the $T - T_g$ then efforts to predict when sticking will occur (during both drying and storage) only need to concentrate on the $T - T_g$ and not the individual temperature and moisture conditions (moisture content and/or water activity) of an amorphous food powder.

## 2. Materials and methods

### 2.1. Materials

Edible grade sucrose (New Zealand Sugar Company Limited, Auckland, New Zealand), grade I maltose monohydrate (Sigma Chemical Company, St Louis, MO), USP grade α-lactose monohydrate (The Lactose Company of New Zealand, Hawera, New Zealand), analytical grade D(+) glucose (BDH Laboratory Supplies, England) and D(+) galactose (UNIVAR, Asia Pacific Specialty Chemicals Limited, Australia) and general purpose reagent D(−) fructose (BDH Laboratory Supplies, England) were used for all experiments. Approximately 20% solutions were made from each powder, which were frozen at −18 °C for at least 20 h and then placed in a −70 °C freezer overnight. The samples were placed into a freeze dryer (Cuddon Freeze Dryers, Blenheim, New Zealand) with a vacuum of 0.01 mbar and a plate temperature of −10 °C. The plate temperature was then increased by 10 °C every 24 h and the samples were removed from the freeze dryer once they had been held at 20 °C for at least 24 h. Ramping the temperature in such a way increased the rate of drying but also prevented retrograde collapse (i.e. collapse of the freeze-dried matrix during warming (MacKenzie, 1975)).

Due to collapse problems associated with freeze drying glucose, galactose and fructose, these amorphous powders were made by stabilising the lower molecular weight sugar with lactose. Solutions of lactose and the lower molecular weight sugar were made in the proportion of (lactose:other sugar) 80:20 for glucose and galactose and 90:10 for fructose. This elevated the $T_g$ for the powder, thereby making it easier to freeze dry and also allowing a larger range of RH and temperature conditions to be tested. The resulting powders were easier to test given the existing rig set-up (see details below) where temperatures less than ambient could not be achieved. Given the dry $T_g$ values for amorphous glucose, galactose and fructose (Roos, 1993), temperatures less than ambient conditions (~20 °C) would need to be used for stickiness trials on the pure sugars. The freeze-dried two component powders were then examined using a polarising microscope to determine whether any crystals were present. Crystals were seen in all mixture powders although it was not possible to determine whether these were crystals of lactose, the lower molecular weight sugar or both.

### 2.2. Differential scanning calorimetry

$T_g$ versus water activity ($a_w$) profiles were only measured for the two component powders. Samples of each powder were placed over saturated salt solutions of lithium chloride, potassium acetate and magnesium chloride to give RH environments of 11%, 23% and 33%, respectively, at 20 °C (Greenspan, 1977). Phosphorous pentoxide was used to give a RH of 0%. A differential scanning calorimeter (DSC: Perkin–Elmer DSC-7, The Perkin–Elmer Corporation, Norwalk, CT) was used to determine the $T_g$. The temperature calibration was performed using the melting points of indium (156.6 °C) and deionised water (0 °C). Sealed 20 μl aluminium pans were used for measurements; an empty pan was used as the reference. DSC pans were filled with powder in an environment of either dry air or nitrogen gas to prevent moisture adsorption prior to measurement. Measurements were made in duplicate.

### 2.3. Temperature/relative humidity rig and testing chamber

A rig, described in O'Donnell, Bronlund, Brooks, and Paterson (2002) was used to provide air at a constant RH (±1.2–1.8%), temperature (±0.2–0.3 °C) and flow rate. The air stream was then directed into either a fluidised bed arrangement or the testing chamber. The fluidised bed was used for preconditioning the powder prior to experimentation and the testing chamber was used to perform cohesion trials. The testing chamber consisted of a glass enclosure, segmented distributor plate and a blow tester. The distributor plate, which the powder was placed upon, was segmented so that discrete measurements of powder cohesiveness could be taken without disturbing the entire powder bed. The blow tester was used to provide a measurement of powder cohesiveness. Full details of the testing chamber and blow tester are given in Paterson et al. (2001) and Foster, Bronlund, and Paterson (2005). The blow tester operates by blowing air (at ambient RH and temperature conditions) from a compressed air line, over the powder bed. The flow rate of the air is increased with time until the surface of the powder bed is disturbed and a channel is blown in the powder bed. As the powder becomes more cohesive, the flow rate of air required to blow a channel in the bed increases. Each caking strength measurement took between 10 and 30 s to complete and, given the flow rate of conditioned air continuously entering the testing chamber (~4 l $\text{min}^{-1}$), this did not appear to adversely affect the temperature and RH conditions within the testing chamber.

### 2.4. Preconditioning of the powders

The freeze dried powder was preconditioned, using the temperature/RH rig, to the RH that the cohesion experiments were to be carried out at. This was done by fluidising the powder at a given RH for approximately 15 h. Care was taken to ensure that the water activity of the powder would render the powder stable at ambient temperatures, i.e. under the powder's $T_g$, so that sticking or crystallisation did not occur prior to commencing the cohesion trials. The powder was preconditioned, in batches, to a number of water activities, so that the cohesion trials could be carried out using a variety of temperature and RH conditions to achieve similar $T - T_g$ values for the powder. After preconditioning, the powder was stored in airtight plastic bags in a 4 °C room until use. Each plastic bag contained enough powder to perform one experiment.

### 2.5. $T - T_g$ cohesion trials

The temperature/RH rig was used to provide the appropriate testing conditions by adjusting the incoming pressure, outlet pressure, temperature of water in the columns and the heater temperature as described in O'Donnell et al. (2002). Once the desired RH and temperature conditions were achieved in the testing chamber, the preconditioned powder was poured into the segments of the testing plate (see Foster et al., 2005). The powder was then levelled off using a sharp edge. Care was taken to use the same procedure for filling the segmented distributor plate with powder each time an experiment was performed. This is likely to have kept the thickness, density and uniformity of the powder bed approximately constant with each experiment although this was not measured. The glass lid of the chamber was fitted and timing commenced. Temperature and RH conditions in the testing chamber and the powder strength, as measured using the blow tester (Paterson et al., 2001), were measured with time. Measurement concluded when the powder strength became constant, the powder strength exceeded the maximum reading obtainable with the equipment used, or the maximum number of readings for the set-up had been taken. The RH used during testing was generally within ±2% of the RH that the powder had been preconditioned to. In this way, the powder was only exposed to a step change in temperature at the start of each experiment. A slight difference between the powder RH and that being tested was not considered important, as the surface of the powder would equilibrate very quickly to the conditions being tested. Chatterjee (2004) and Zuo (2004) found that an exposure time of 0.05 s to appropriate air conditions was sufficient to affect the surface stickiness of dairy powders. Since sticking is a surface related phenomenon, it is only important that the surface is at the temperature and RH conditions that are being tested.

## 3. Results and discussion

The results and analyses performed on the data from the cohesion trials are discussed using amorphous sucrose as the example. It is noted that the same analysis was used for all amorphous sugars. The only exception is that the analysis of the data for amorphous glucose/lactose, galactose/lactose and fructose/lactose powders used the actual $T_g$ versus $a_w$ profiles measured for the blends. The analysis for amorphous sucrose and maltose used the $T_g$ profiles for the sugars given in Foster (2002) which were determined using data from Finegold, Franks, and Hatley (1989), Orford, Parker, Ring, and Smith (1989), Orford, Parker, and Ring (1990), Roos and Karel (1990), Izzard, Ablett, and Lillford (1991), Roos and Karel (1991a, 1991b, 1991c, 1991d), Roos (1993), van den Dries, van Dusschoten, and Hemminga (1998), Miller and de Pablo (2000), Noel, Parker, and Ring (2000) and Thielmann and Williams (2000). The $T_g$ versus $a_w$ profiles for each powder are given in Fig. 1. Using the RH and temperature conditions that the powder was exposed to during the cohesion trial and the $T_g$ versus $a_w$ profile for the powder, the $T - T_g$ for that trial was calculated.

Fig. 2 shows a collection of trials performed using amorphous sucrose with the RH and temperature conditions and consequent $T - T_g$ given in the legend. It can be seen that for low $T - T_g$ values, i.e. a $T - T_g \approx 10$ °C, the change in cohesiveness with time is slow and only a small change can be seen over a period of 16 h. In fact, these trials ran for up to 35 h ($\sim$1.5 days) and the change in cohesiveness over that time was 2 l min$^{-1}$, which did not result in a noticeably sticky or lumpy powder. $T - T_g$ values of 16–22 °C gave much higher changes in cohesiveness with time. In general, it can be seen that the rate of change in powder cohesiveness with time increases as the $T - T_g$ increases. $T - T_g$ values greater than 22 °C for amorphous sucrose were required for sticking to occur almost instantaneously. Fig. 2 also shows that trials with similar $T - T_g$ values gave similar rates of cohesion even when the RH and temperature conditions used to obtain a particular $T - T_g$ differed. This can be seen more clearly in Fig. 3 which plots the rate of change in powder cohesiveness with time versus the $T - T_g$. This was obtained by taking the slopes of the data (caking strength (l min$^{-1}$) versus time (h)) in Fig. 2 and plotting against their respective $T - T_g$ values. In some cases, where the data did not have a linear relationship, the slope was taken from the initial change in powder cohesiveness. Referring to Fig. 2, a nonlinear relationship can be seen for the trial performed with RH and temperature conditions of 21.4% and 42.7 °C, respectively, giving a $T - T_g$ of 14.5 °C. This data shows an initial linear change in powder cohesiveness and then the rate decreases and the curve eventually plateaus. This kind of relationship can be explained by considering the relative rates of flow and crystallisation in the powder at those conditions. Initially, flow would occur and liquid bridges would form giving the initial increase in the cohesive strength of the powder. At some point in time, crystallisation would begin. As crystallisation proceeded the rubbery amorphous material that was forming the liquid bridges would begin to get fixed in place as some of the sugar molecules began to align with each other and crystallise. The change in cohesiveness with time would then occur at a different rate, which would be more proportional to the rate of crystallisation, as the formation of liquid bridges slowed down and the solidification of the liquid bridges took place. This would show up as a curved line in Fig. 2 if the rate of sticking and rate of crystallisation/caking differed. Where this type of relationship has occurred, the initial rate data which is likely to be proportional to the rate of sticking was used. Roos and Karel (1992) showed that $\sim$20% crystallisation of amorphous lactose, at a constant $T - T_g$ value of 16.5 °C, occurs within 24 h. It was also found that the rate of crystallisation was fairly constant until $\sim$20% crystallinity, after which, the rate increased exponentially.

Fig. 3 shows that the rate of cohesion is related to the $T - T_g$ of the powder and not the individual RH/$a_w$ and temperature conditions used to obtain a specific $T - T_g$ value. It also shows that the rate of cohesion increases proportionally with an increase in the $T - T_g$ value. The rate of sticking has been graphed on a log scale due to the large difference in rates for low and high $T - T_g$ values. Using the log scale, a fairly linear relationship can be seen

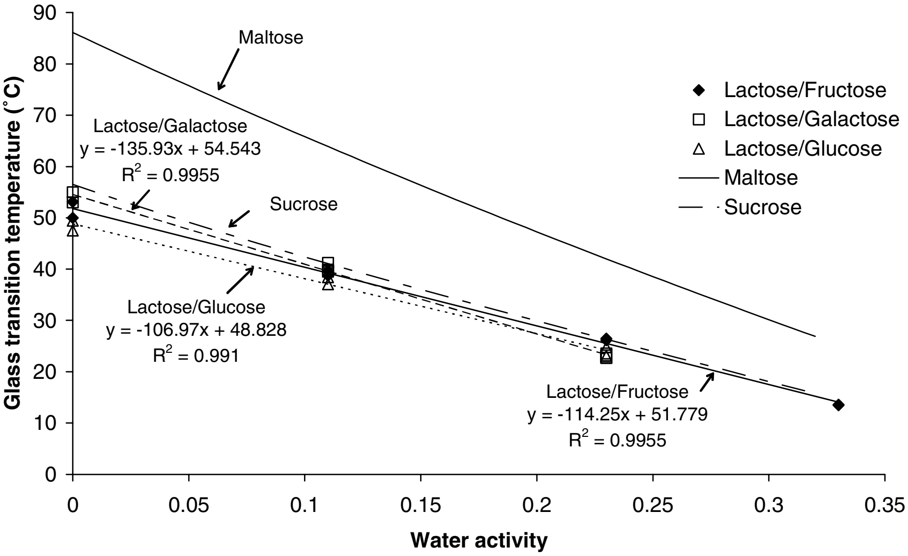

Fig. 1. Glass transition temperature versus water activity profiles for amorphous maltose, sucrose, lactose/glucose, lactose/galactose and lactose/fructose powders.

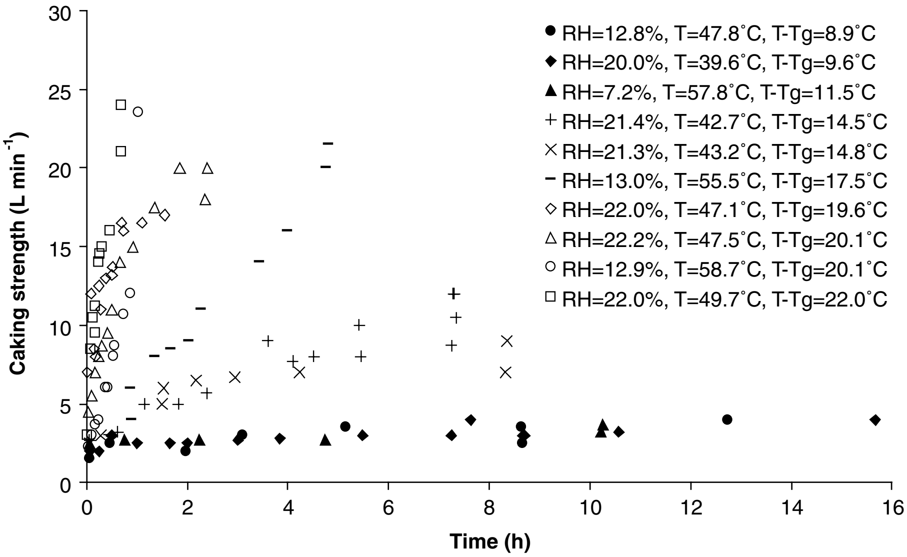

Fig. 2. Caking strength versus time data for amorphous sucrose powder.

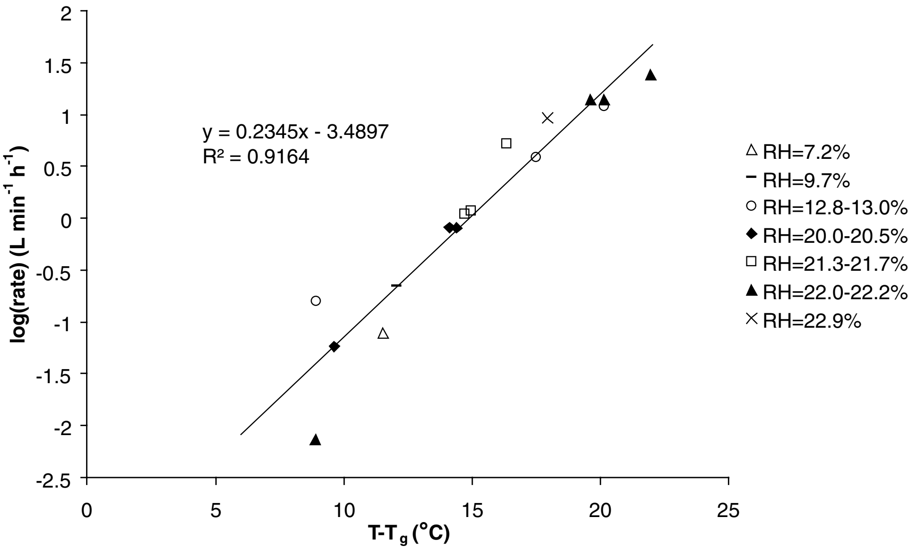

Fig. 3. Rate of cohesiveness development versus $T - T_g$ for amorphous sucrose powder.

between the log of the rate of sticking and the $T - T_g$ of the powder.

The same analysis was used for amorphous maltose and amorphous glucose/lactose, galactose/lactose and fructose/lactose mixtures (see Figs. 4–7). Linear regression was used to fit a straight line to the data. In the case of amorphous glucose/lactose (Fig. 5), the very slow rates were excluded from the regression analysis as these rates were indistinguishable from zero. It would require a certain $T - T_g$ before the rate of stickiness development was distinguishable from zero, therefore a relationship can only be fitted from that $T - T_g$ and above. This was not observed for the other amorphous sugar powders. Some data at very high $T - T_g$ values appeared to be outliers and were excluded from the analysis (circled data in Fig. 5), as a lower rate of cohesion was found with very high $T - T_g$ values. Experiments performed at $T - T_g$ values of 35.0 and 37.4 °C showed instantaneous sticking i.e. the maximum caking strength value that could be achieved using the blow tester was exceeded during the time it took to reassemble the testing chamber after commencing the experiment. Collapse was also noted with these experiments indicating that extensive flow was occurring. It is therefore not clear whether these low rates of sticking at high $T - T_g$ values were due to a different mechanism occurring or whether they were outliers due to deterioration of the samples or errors during experimentation.

Figs. 4–7 show the relationship between the rate of cohesion and the $T - T_g$ for amorphous maltose, amorphous glucose/lactose, galactose/lactose and fructose/lactose, respectively. Figs. 4–7 show that the rate of development of cohesiveness in amorphous maltose, and glucose/

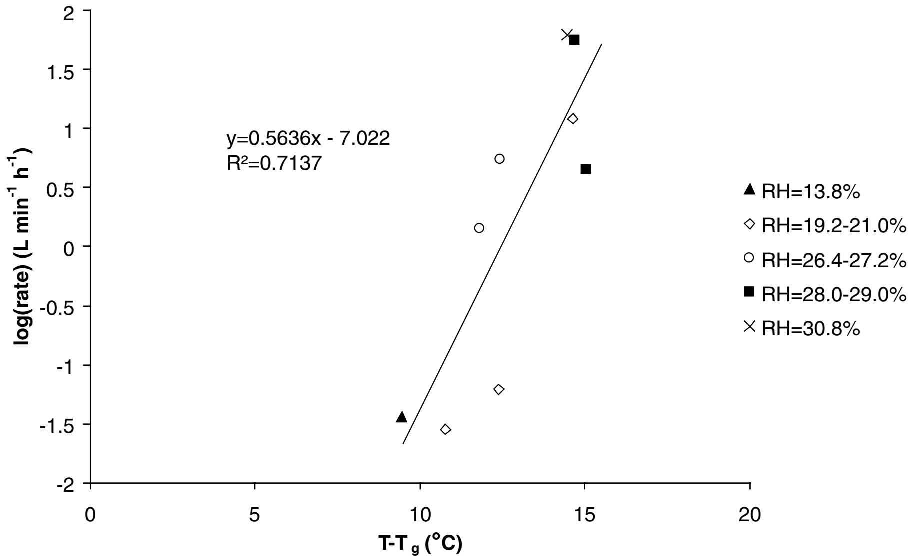

Fig. 4. Rate of cohesiveness development versus $T - T_g$ for amorphous maltose powder.

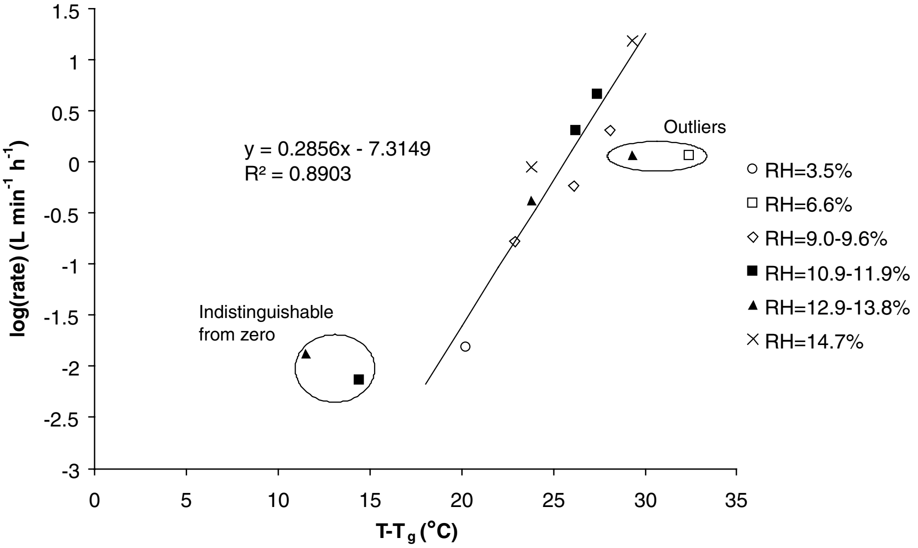

Fig. 5. Rate of cohesiveness development versus $T - T_g$ for amorphous lactose/glucose powder.

lactose, galactose/lactose and fructose/lactose mixtures are proportional to the $T - T_g$ and not the individual temperature and RH conditions used to obtain the particular $T - T_g$. The rate of stickiness development of amorphous lactose has also been found to be proportional to the $T - T_g$ of the powder and not the conditions used to obtain the $T - T_g$ (Paterson et al., 2005).

Table 1 shows the $T - T_g$ values that resulted in instantaneous sticking of the amorphous sugar. Instantaneous sticking occurred when conditions were such that the maximum blow test reading was exceeded in the time taken to reassemble the testing chamber and take the first reading (less than 2 min). The $T - T_g$ values given in Table 1 are not the instantaneous sticking points, they are just values for which instantaneous sticking did occur. It is possible that instantaneous sticking could also occur at a slightly lower $T - T_g$ for a particular sugar. It is noted that instantaneous sticking did not occur under any of the experimental conditions used for amorphous galactose/lactose powder. However, the powder cohesiveness did exceed the maximum value for one experiment in less than 5 min so this $T - T_g$ has been stated in Table 1. It is noted that for the lower molecular weight sugars (i.e. glucose, galactose and fructose) that were made as mixtures with lactose, relatively higher $T - T_g$ values are required for instantaneous sticking. These freeze dried mixtures contained some crystalline material, as observed using a polarising microscope. It is possible that the presence of crystalline material affects the rate of flow (through increased viscosity) and therefore sticking and caking in these powders. The

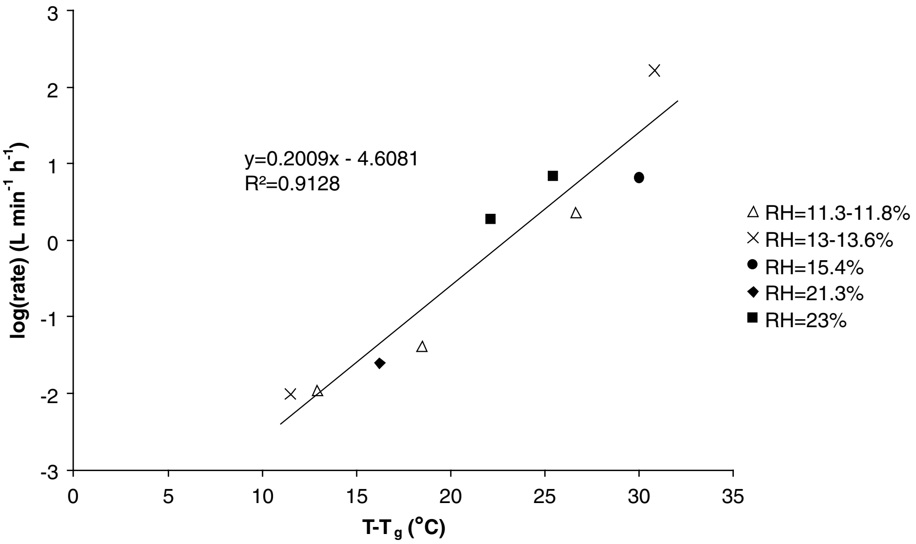

Fig. 6. Rate of cohesiveness development versus $T - T_g$ for amorphous lactose/galactose powder.

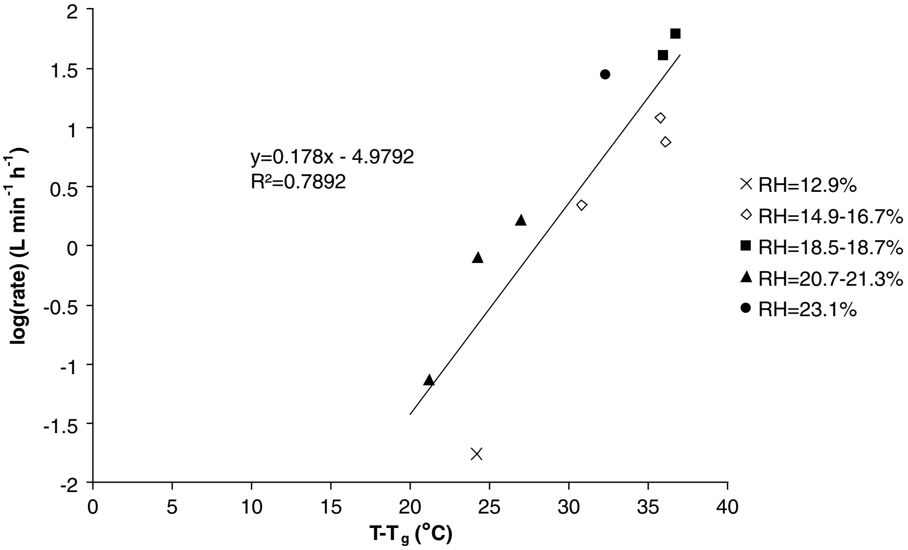

Fig. 7. Rate of cohesiveness development versus $T - T_g$ for amorphous lactose/fructose powder.

Table 1
$T - T_g$ values for instantaneous sticking

| Sugar | $T - T_g$ (°C) |
|---|---|
| Lactose | 25.0[a] |
| Sucrose | 23.0 |
| Maltose | 19.0, 25.3, 29.0 |
| Glucose/Lactose | 35.0, 37.4 |
| Galactose/Lactose | 30.8 |
| Fructose/Lactose | 41.3 |

[a] Brooks (2000).

presence of some crystalline material results in there being less amorphous material available for forming bridges. Therefore, it would take more time for the same level of flow and stickiness to occur since there is less amorphous material available at the contact points between particles. For the same sized amorphous bridge to form, it would require amorphous material from further away to flow to the liquid bridge. As stated in Eq. (1) (Downton et al., 1982), an increase in the distance over which flow must occur (represented by the term $KD$) will decrease the tendency towards sticking and therefore require increased time for the same level of sticking to take place.

$$\mu = \frac{k\sigma t}{KD} \tag{1}$$

where $\mu$ is the viscosity (Pa s), $k$ the dimensionless proportionality constant of order unity: the surface tension (N m$^{-1}$), $t$ is the time (s), $K$ is the fraction of the particle diameter required as a bridge width for a sufficiently strong bridge and $D$ is the particle diameter (m).

An alternative explanation is the effect of the viscosity and/or surface tension of the mixture. Maltini and Anese

(1995) compared the viscosities of concentrated glucose, fructose and sucrose solutions with those predicted using the Williams–Landel–Ferry equation (Williams, Landel, & Ferry, 1955). There did not appear to be any significant difference in the experimental and predicted viscosities when all comparisons used the universal constants and the viscosity for the glass was assumed to be $10^{14}$ cP. Therefore, there was no difference in viscosity between the different sugars when related to the $T - T_g$. Mathlouthi, Hutleau, and Angiboust (1996) also found there to be very little difference between the viscosities of sucrose, glucose and fructose solutions. Downton et al. (1982) stated that the rate at which the bridges are formed is governed by the viscosity of the material. With this in mind, there should be no difference between the rate of bridge formation for the different sugars (if surface tension is constant) since the viscosities appear to be the same when related to the $T - T_g$. The driving force for the formation of bridges is governed by the surface tension of the amorphous material, i.e. as the surface tension increases, the tendency towards sticking increases. Comparing the surface tension of concentrated lactose and 10% sucrose solutions (66 mN m$^{-1}$ (Bronlund, 1997) and 71.5 mN m$^{-1}$ (Mathlouthi et al., 1996), respectively), sucrose has a higher surface tension therefore the $T - T_g$ at which instantaneous sticking occurs should be lower, since the driving force for bridge formation is higher. This is seen to be the case in Table 1. The surface tensions of 10% fructose and glucose solutions, as found by Mathlouthi et al. (1996), are 73.8 and 72.8 mN m$^{-1}$, respectively. The surface tensions for these sugars are greater than that for sucrose. However, a decrease in the $T - T_g$ required for instantaneous sticking was not found for these sugars when mixed with lactose. The $T - T_g$ for instantaneous sticking was higher than that required for amorphous lactose. This indicates that the difference in the $T - T_g$ values for instantaneous sticking is not related to the surface tension or viscosity of the different sugars. It is more likely that the difference is due to the presence of crystalline material, which would increase the viscosity of the sugar mixtures and require amorphous sugar from further away to flow and form liquid bridges.

At this point, it is worthwhile addressing some factors that would affect the results obtained. Firstly, the size and shape of the powder particles were not measured. Although it is probable that the particle size and shape differed between different types of powder (e.g. sucrose, maltose, glucose/lactose, galactose/lactose and fructose/lactose), these characteristics are likely to have been the same within each powder as only one batch of each amorphous sugar powder was made and all experiments were performed using that same batch of powder. Carrying out all experiments on the same batch of powder also eliminated problems such as having two different batches of sucrose, for example, with different amorphous sucrose contents. Care was taken with the storage of all powders before experimentation, therefore, different experiments on the same sugar should have been performed using a sugar powder of the same amorphous sugar content and the same particle characteristics. It is important to note that the results obtained from this work are significant because of the trends that the data show, rather than the absolute values.

This work supports previous work (e.g., Levine & Slade, 1986; Özkan, Withy, & Chen, 2003; Paterson et al., 2005; Roos & Karel, 1992) which has shown cohesion to be a time-dependent phenomenon. That is, the rate that cohesion occurs will depend on the driving force for cohesion, which is the $T - T_g$ (representing a decrease in viscosity) for the powder, and not the actual temperature and RH conditions being used. The experimental procedure developed by Paterson et al. (2001) and used in this work contributes to research into glass transition related flowability problems as it allows readings to be taken over time rather than just one point being measured. This allows the observation of the time-dependent side of these sticking and caking problems to be made under constant $T - T_g$ (temperature and RH) conditions. The variable of time provides useful information when dealing with stickiness and caking problems during fluidised bed drying and storage where longer residence times are experienced.

As cohesion is time dependent, a certain $T - T_g$ will be required before instantaneous stickiness will occur. Typical sticky point curves which have been measured using very short time frames should show that instantaneous sticking occurs at a constant amount above the $T_g$. This has been demonstrated by Hennigs et al. (2001) and Ozmen and Langrish (2002) for skim milk powder. Recently, it was concluded that the sticky point temperature was not too dissimilar to the $T_g$ and that the $T_g$ (as measured by DSC) could be used as the sticky point temperature in skim milk powder (Ozmen & Langrish, 2002). This work was incorrect in stating that the $T_g$ could be used as the sticky point temperature, due to closeness of the two values, as the sticky point may be considerably higher than the $T_g$, e.g. 23.3 °C (Hennigs et al., 2001), and even Ozmen and Langrish's (2002) work showed it to be 14–22 °C higher than the $T_g$. The work presented in this paper has shown that amorphous sugars require a certain $T - T_g$ (e.g. >19 °C for amorphous maltose) before stickiness begins to occur in a short time. Therefore, although it may be useful to use a typical "sticky point temperature" measurement during situations such as spray drying where the residence time is short, it is not useful to use the $T_g$ as the sticky point in these situations as it is overly conservative.

## 4. Conclusions

This work confirmed that the rate of change in cohesiveness of amorphous sugars (sucrose, maltose, glucose, galactose and fructose) is related to the $T - T_g$ of the powder and not the individual temperature and RH conditions used to obtain a particular $T - T_g$. Different combinations of temperature and RH conditions which yield the same $T - T_g$ will result in the same rate of cohesion between particles. The time-dependent nature of stickiness and caking has been demonstrated and shows that the same level of cohesiveness can be obtained using different $T - T_g$ values, so long as the appropriate amount of time is given in each case for the bridges to form.

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