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# The physical nature of stickiness in the spray drying of dairy products—a review

D. J. O’Callaghan · S. A. Hogan

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**Abstract** This paper reviews developments in understanding the issue of stickiness in the spray drying of dairy products, with particular reference to the physical basis of stickiness. Investigation of stickiness in spray drying has followed two main directions: (1) approaches to measuring the phenomena of stickiness by empirical means and (2) understanding stickiness in the context of the science of soft matter (e.g. as being related to glass transition phenomena). The former approach has led to several techniques that have been used as empirical tools to study the influence of product formulation on stickiness. The latter approach underpins applied research in this area with a body of knowledge being developed in materials science. Empirical-type measurements can be used to validate mechanistic models which are emerging. Tools in the field of microscopy, such as X-ray photon spectroscopy, energy-dispersive X-ray spectroscopy and atomic force microscopy, are beginning to contribute to a better understanding of the underlying physical phenomena.

**Keywords** Stickiness · Spray drying · Dairy products

## 1 Introduction

When spray drying technology is used to manufacture food products, powder handling difficulties can arise for some products due to adhesion to equipment surfaces. Under some conditions, powder adheres to the surface of the drying chamber, usually near exit points, leading to possible blockage, downtime and loss of product (Ozmen and Langrish 2003). This is generally considered to result from the formation of liquid bridges containing amorphous sugar or other dissolved material conferring high surface tension, or perhaps lipid material in some cases, between particles and equipment surfaces (Palzer 2005; Foster et al. 2006). Investigation of stickiness in

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spray drying has followed two main directions: (1) approaches to measuring the phenomenon of stickiness and (2) understanding stickiness in the context of the science of soft matter (e.g. as being related to glass transition phenomena). The former approach has led to several techniques that have been used as empirical tools to study the influence of product formulation on stickiness. The latter approach underpins applied research in this area with a body of knowledge being developed in materials science, coupled with the theoretical framework of molecular dynamics.

As the phenomenon of stickiness is not itself a fundamental property of a material, several empirical approaches have been investigated, some of which mimic the spray drying process. The understanding of stickiness in the spray drying of foods has improved through combining these approaches.

## 2 Stickiness as a physical phenomenon in spray drying

### 2.1 Glass transition phenomena

For powders with a high carbohydrate content, the phenomenon of stickiness is related to the physical state of the carbohydrate components, i.e. normally an amorphous glassy state. In the course of drying, viscosity and surface tension become extremely high around a critical water activity level which is dependent on composition and temperature (Adhikari et al. 2001, 2007; Palzer 2005; Foster et al. 2006; Roos 2009; Hogan and O'Callaghan 2010). The high viscosity/high surface tension state is sometimes referred to as a rubbery state (Roos and Karel 1992). Further drying leads to a solid glassy state which is non-sticky (Foster et al. 2006). There is a region roughly between the rubbery and non-sticky states known as a glass transition region (Brostow et al. 2008). Stickiness problems in spray drying are related to the powder coming in contact with equipment surfaces whilst in the glass transition state, sometimes referred to as a plastic state (Bhandari and Howes 1999). This region is associated with changes in energy state at the molecular level which affect the geometrical ordering of molecules, the microstructure and flow properties. Several physical manifestations of changes in thermoplastic properties can be observed as a material passes through such a transition:

1. A discontinuity in specific heat capacity, i.e. the rate of heat absorption (or desorption) with respect to temperature changes (i.e.  $dQ/dT$  is not constant)
2. Large changes in viscoelastic properties with respect to temperature, as described by empirical equations, e.g. the Williams–Landel–Ferry (WLF) equation

$$\log_{10} \frac{\eta}{\eta_g} = \frac{-C_1(T - T_g)}{C_2 + (T - T_g)}$$

where  $\eta$  is viscosity at temperature  $T$ ,  $\eta_g$  the viscosity at glass transition temperature  $T_g$ , and  $C_1$  and  $C_2$  are constants (Bhandari and Howes 1999; Adhikari et al. 2001; Abiad et al. 2009). This equation can be used to describe viscosity for temperatures between  $T_g$  and  $T_g + 100$  °C.

3. Large changes in dielectric properties (electrical permittivity and dielectric loss) and molecular relaxation characteristics, which are related to the time required to align dipoles, as functions of temperature (Silalai and Roos 2011)

4. A change in coefficient of thermal expansion of the material, which can be observed as a contraction or expansion in volume under compression as temperature changes (Boonyai et al. 2005; Rahman et al. 2007)
5. Changes in surface tension with respect to temperature

These phenomena are interrelated by the fact that (1) heat capacity, which is a measure of energy required for molecular vibration, and (2) viscoelastic and dielectric properties are related to the space restriction on molecules, intermolecular forces, volume fraction and material density (Meste et al. 2002). A clue to the implication of the coefficient of expansion for surface tension and, hence, stickiness is provided by Sauer and Dee (2002) who showed that, for synthetic polymers, melt surface tension is intrinsically related to the pressure–volume–temperature state.

## 2.2 Principles of measurement of state changes near glass transition

A wide range of measurement techniques are available for detecting the changes described above (Abiad et al. 2009). Commonly used techniques are (1) differential scanning calorimetry (DSC), (2) dynamic mechanical analysis (DMA), (3) dielectric analysis (DEA) and (4) thermomechanical analysis. Surface tension is relatively difficult to measure in semi-solid materials, as opposed to liquids, and tends not to be used as a direct measure of powder stickiness. The main features of the most common techniques for investigating glass transition can be summarised as follows.

### 2.2.1 DSC analysis

Glass transition temperature is most commonly defined in relation to DSC measurement using hermetically sealed pans to maintain constant moisture level. It is important to note that glass transition does not occur at a precise temperature or point on a state diagram, but rather over a region, the extent of which depends on dynamic changes taking place during the measurement, e.g. rate of heating, cooling or water evaporation (Roos 1993; Rahman et al. 2007). In DSC analysis, three glass transition parameters are frequently defined, namely an onset temperature,  $T_{gi}$ , where a change in specific heat capacity is detected as temperature is increased; a midpoint temperature,  $T_m$ ; and an endset temperature,  $T_{ge}$ . The onset and endset temperatures,  $T_{gi}$  and  $T_{ge}$ , are determined graphically by intersection of tangents;  $T_m$  is determined as a point of inflection on a plot of heat flow with respect to temperature. For useful scientific discussion, and to compare results of different studies, a  $T_g$  value should specify the measurement method (including heating rate) and definition of  $T_g$  (Kasapis et al. 2003). For example, a good practice is to refer to “onset temperature of glass transition” (Silalai and Roos 2011). Syamaladevi et al. (2012) found that for three common carbohydrates, the difference between  $T_{ge}$  and  $T_{gi}$  was in the range 4–15 °C at a typical heating rate of 3 °C/min.

### 2.2.2 Dynamic mechanical analysis

The application of the glass transition concept implies that a powder has some viscoelastic properties. Changes in the viscoelastic properties of powders are

determined from the elastic and loss moduli obtained under low strain harmonic shear motion of a sealed pressed sample at defined oscillation frequencies, typically in the range 0.1–20 Hz, on dedicated DMA instruments or using precision rheometers (Royall et al. 2005; Rahman et al. 2007; Hogan et al. 2010). Kasapis et al. (2003) showed that the frequency of oscillation needs to be taken into account in DMA, just as the heating rate needs to be taken into account in DSC analysis; indeed, a spectrum of measurements (across a wide frequency range) is normally analysed. A definition of mechanical glass transition for food systems was derived (Kasapis et al. 2003).

### 2.2.3 Dielectric spectroscopy

Dielectric spectroscopy measures changes in dielectric properties detected by the energy response to an oscillating electric field, typically in the audio frequency range, using DEA instruments.

DMA and DEA measurements are often analysed using relaxation times to determine an  $\alpha$  relaxation temperature,  $T_\alpha$  (Silalai and Roos 2011).

The concept of relaxation arises in relation to glass transition due to the fact that material in the glass transition region behaves like a supercooled melt, which is a metastable state that would, given sufficient time, relax into a stable (crystalline) state. Indeed, the WLF equation can be used to predict crystallisation time (Langrish 2008). Since a theoretical relaxation time can be calculated from oscillatory measurements on the basis of simple viscoelastic behaviour (the dashpot spring or Kelvin–Voigt model), viscosity or elastic modulus may be conveniently transformed into a relaxation time parameter, and the sudden changes in this parameter that take place in the glass transition region can be used to determine  $T_\alpha$  (Champion et al. 2000). An analogous approach can be adopted using nuclear magnetic resonance (NMR; Stuergea 2006). Lloyd et al. (1996) compared  $T_g$  (onset temperature) using DSC with a glass transition temperature based on NMR spin relaxation measurement for spray-dried amorphous lactose and found agreement within  $\pm 10$  °C.

It follows from the foregoing that the empirical equations used to model viscosity (e.g. the WLF equation) may, with appropriate substitutions, be used to model relaxation time, i.e. relaxation time can in general be taken as viscosity on some other scale. The most common equations cited in the literature for modelling viscosity or relaxation time at temperatures above  $T_g$  (and up to  $T_g + 100$  °C) are the WLF equation mentioned earlier and the Vogel–Fulcher–Tammann (VFT) equation (also known as the Vogel–Tammann–Fulcher equation).

$$\eta = \eta_\infty \exp\left(\frac{D T_0}{T - T_0}\right)$$

where  $\eta_\infty$ ,  $D$ , and  $T_0$  are constants.  $\eta_\infty$  is a supposed viscosity at  $T = \infty$ .  $T_0$  is usually several degree Celsius below  $T_g$ . Even though the coefficients of the WLF and VFT equations have quite different interpretations, the equations are mathematically equivalent and, therefore, interconvertible (Kopesky et al. 2006).

In the literature on the physical properties of polymers in the glass transition region, attempts have been made to define a parameter, fragility, as an index of departure from Arrhenian behaviour, i.e. where glass transition is seen as a departure from the Arrhenius relationship with temperature for some parameter. A material that

only deviates slightly from Arrhenian behaviour is said to be “strong”, whilst a material that deviates considerably is said to be “fragile”. Arrhenian behaviour in this context means a viscosity/temperature relationship of the form

$$\log_{10} \eta = a + b \left( \frac{T_g}{T} \right)$$

where  $T_g$  and  $T$  are in absolute units and  $a$  and  $b$  are constants. The more curved a semi-log plot of viscosity (or of relaxation time) of a material versus  $T_g/T$  is, the more fragile the material is said to be (Angell 2002). The fragility concept has not as yet been proven useful in the analysis of food polymers as different definitions and methods of determination lead to very different results (Syamaladevi et al. 2012).

The relaxation time concept is also of interest because of its direct linkage to molecular dynamics. Molecular dynamics simulation software has been used to study the temperature-dependent relaxation time for individual proteins, and it was shown that the simulation results could be fitted to the VFT equation (Baysal and Atilgan 2005).

In practice, glass transition/alpha transition is a useful concept in the discussion of the stickiness of carbohydrate-rich food systems as it can relate food composition to conditions at which stickiness is observed (Roos 2002). This was confirmed by Bhandari et al. (1997a) who developed a semi-empirical model for the spray drying of sugar-rich foods and showed that (1) the relative drying indices for various anhydrous sugars and maltodextrin had a linear relationship with  $T_g$  and that (2) the relative drying indices for a range of pineapple juice/maltodextrin blends also had a linear relationship with  $T_g$  over a range of water activity from 0.085 to 0.120.

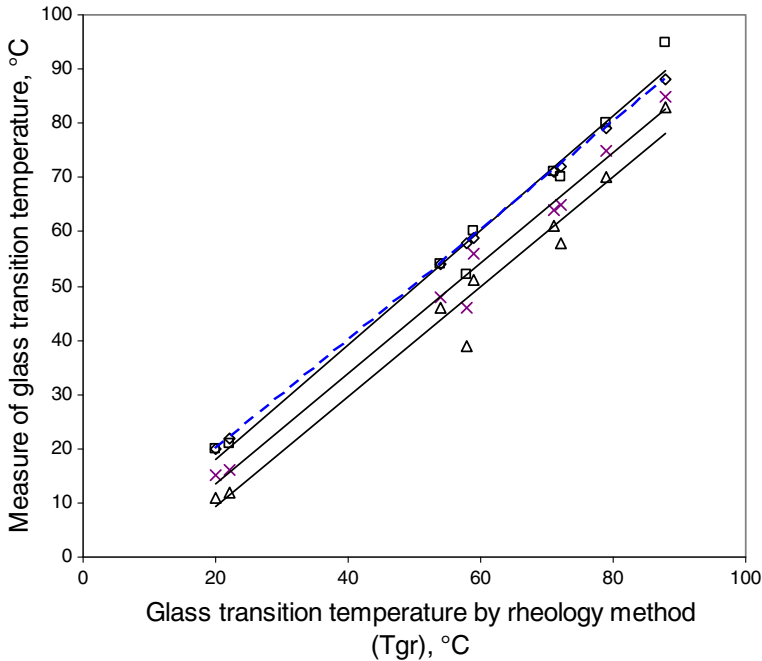
In general,  $T_g$  and  $T_\alpha$  are dependent on the water activity level as well as on the composition. Stickiness is exhibited when the temperature exceeds  $T_g$  by some increment that is determined experimentally (Adhikari et al. 2001; Paterson et al. 2005; Hogan and O’Callaghan 2010).

#### 2.2.4 Thermal mechanical compression

Changes in the coefficient of thermal expansion in the glass transition region have been exploited as a means of determining a glass transition temperature (Boonyai et al. 2007). Such a technique was adapted by Hogan et al. (2010) as a rheological method for determining a glass transition temperature, denoted  $T_{gr}$ . The method was tested on a range of non-fat dairy powders with a wide range of protein and lactose levels and casein/whey protein ratios and gave a range of  $T_{gr}$  values close to the  $T_{ge}$  values ( $\pm 3$  °C) and above the  $T_{gi}$  values (by  $10 \pm 4$  °C) for the same powders (Fig. 1). This has become a standard laboratory method (Schuck et al. 2012).

#### 2.2.5 Other techniques cited for $T_g$ measurement

Other techniques used for determining  $T_g$  include positron annihilation lifetime spectroscopy, oscillatory squeezing flow, inverse gas chromatography, scanning probe microscopy (e.g. atomic force microscopy) and thermally stimulated depolarizing current (Abiad et al. 2009).



**Fig. 1** Comparison of  $T_g$  measurement by DSC ( $T_{gi}$ , triangles;  $T_{gm}$ , crosses; and  $T_{ge}$ , squares) and thermomechanical analysis ( $T_{gr}$ , dashed line). Trend lines (solid line) for  $T_{gi}$ ,  $T_{gm}$  and  $T_{ge}$  are shown in order from bottom up. As  $T_{gr}$  data are plotted against themselves, these are in a straight line, for reference. Data from Hogan et al. (2010)

### 2.3 Measurement of stickiness

Stickiness as an issue in spray drying is typically observed as a buildup of powder deposit at the outlet of the spray drying chamber, cyclone blockage or collapse of powder in fluid beds. A wide range of objective techniques for stickiness measurement have been cited (Table 1). Some attempts at quantifying stickiness and investigating the factors that influence it have been made by direct observation of wall deposition or fluid bed collapse either in commercial or pilot-scale plants (Hoffmann 2000; Ozmen and Langrish 2003). Most techniques for stickiness measurement involve either a measurement of flow or adhesion of powder under conditions of controlled temperature and humidity, the latter being achieved in early sticky-point testers by relying on the moisture contained in the powder to equilibrate with surrounding air or, in later techniques, by exposing the powder to a supply of conditioned air. In some techniques, most notably the particle gun, the particle impact velocity can be controlled.

The results obtained from different techniques have been compared with each other and with glass transition measurements and have provided useful information relating stickiness to process conditions, e.g. the influence of air humidity on sticking behaviour. Where stickiness is governed by the carbohydrate content, sticking is in general observed at temperatures above glass transition, and the results obtained using different techniques are in general comparable, with differences between techniques attributable to impact velocities or pressures or contact times (Murti et al. 2010).

**Table 1** Techniques for stickiness measurement

Technique	Conditioned air	Controlled particle velocity	Reference
Wall deposition	No (in situ test)	No (in situ test)	Bhandari et al. (1997a); Ozmen and Langrish (2003)
Shear cell	No	No	Adhikari et al. (2001)
Sticky point powder stirrer methods	No, but equilibrated using sealed apparatus	No	Hennigs et al. (2001)
Mechanically stirred with controlled RH	Yes	No	Kudra (2003)
Optical probe method	No, but equilibrated using sealed apparatus	No	Lockemann (1999); Adhikari et al. (2001)
Cyclone stickiness test	Yes	No	Boonyai et al. (2006); Hogan et al. (2009); Silalai et al. (2009); Intipunya et al. (2009); Hogan and O'Callaghan (2010)
Fluidised bed with stirrer	Yes	No	Kockel et al. (2002)
Micro-fluidised bed	Yes	No	Hogan et al. (2009); Werner et al. (2006); Verdurmen et al. (2006); Murti et al. (2010)
Thermal mechanical compression test	No	No	Boonyai et al. (2005; 2007); Hogan et al. (2010);
Particle gun	Yes	Yes	Paterson et al. (2007a, b); Zuo et al. (2007)
Blow test apparatus	No	No	Boonyai et al. (2004)
Probe tack test <sup>a</sup>	No, but drying conditions are partially simulated	No	Werner et al. (2007a, b)

<sup>a</sup> Measures stickiness in the course of drying a droplet of liquid

## 2.4 Modelling of stickiness

Palzer (2005) developed a mathematical model for the agglomeration of wet particles in a humid environment by liquid bridges and sintering. The model involves adhesive forces due to surface tension leading to liquid bridges which become sinter bridges after a minimum contact time; he fitted this model to a previously cited work. However, particle velocity was not directly included in this model, and Murti et al. (2010) cast some doubt on the applicability of the model to high-velocity situations. Woo et al. (2010) developed a model for wall-particle collisions in which a particle is modelled as a sphere of viscoelastic material that deforms upon colliding with a surface. Using a numerical method, the deformation and trajectory of a particle are modelled, based on its impact velocity, size and viscoelastic parameters (which can be related to temperature vis-à-vis glass transition), to determine whether a particle will adhere or rebound. The model predicts that particles stick at some temperature above  $T_g$ , with low-velocity particles sticking more easily. It would be interesting to see such models being validated experimentally.



### 3 Influence of food components and their concentrations on the surface

#### 3.1 Effect of carbohydrate

Stickiness in general involves changes in the thermodynamic state which are different in nature for sugars, proteins and fats. It has been well established that sugars have a greater tendency to become sticky and have lower glass transition temperatures as their molecular weight decreases, which makes fructose juices much more difficult to dry than dairy formulations containing lactose (Bhandari and Howes 1999; Roos 2002). Similar effects of molecular weight were found for other carbohydrates, such as maltodextrin, although the “baseline” is different for different types of molecule. In all cases, the plasticisation effect of water depresses  $T_g$  and powders become prone to stickiness at temperatures above the  $T_g$  level, which is a moving target where water content or water activity is changing, as in the course of drying (Abiad et al. 2009).

#### 3.2 Effect of protein

Proteins have a sticky, rubbery state around a point where they become “jammed” due to concentration. Support for this statement comes from the fact that colloidal glass transitions can be observed in a concentrated sterically stabilised colloidal system (Weeks et al. 2000). Thus, proteins are very viscous in a liquid concentrate form, but at later stages of drying the stickiness of protein-containing dispersions tends to be dominated by the carbohydrate components. Nevertheless, there are protein–carbohydrate interactions whereby the susceptibility to stickiness in protein/lactose blends is moderated by the presence of protein, becoming less sticky than would be predicted by the effect of glass transition alone (Adhikari et al. 2009; Hogan et al. 2009; Jayasundera et al. 2009 Hogan and O'Callaghan 2010; Silalai and Roos 2010).

#### 3.3 Contribution of fat

Whilst for carbohydrates stickiness is exhibited in a rubbery state, which is strongly influenced by moisture content and temperature, the kind of stickiness associated with fats usually involves melting behaviour (Paterson et al. 2007a). Whole milk powder, fat-filled powder and other formulations containing fat as a major component (e.g. cheese powders with fat content up to 40%) are manufactured by spray drying. Normally, the fat is encapsulated in the liquid concentrate in the form of an emulsion, where proteins act as natural encapsulants due to their surface active properties (Fältdt and Bergenstahl 1994, 1996; Vega and Roos 2006). However, because dehydration in spray drying occurs at air–liquid or air–solid interfaces, any fat molecules that are not encapsulated tend to occupy the outer surfaces of powder particles because of their hydrophobic nature at the expense of proteins or carbohydrates (Nijdam and Langrish 2006). X-ray photon spectroscopy (XPS) analysis suggests that skim milk powder (SMP) and whole milk powder (WMP) have thin layers of fat on their surfaces, but this does not normally cause a problem with powder handling.

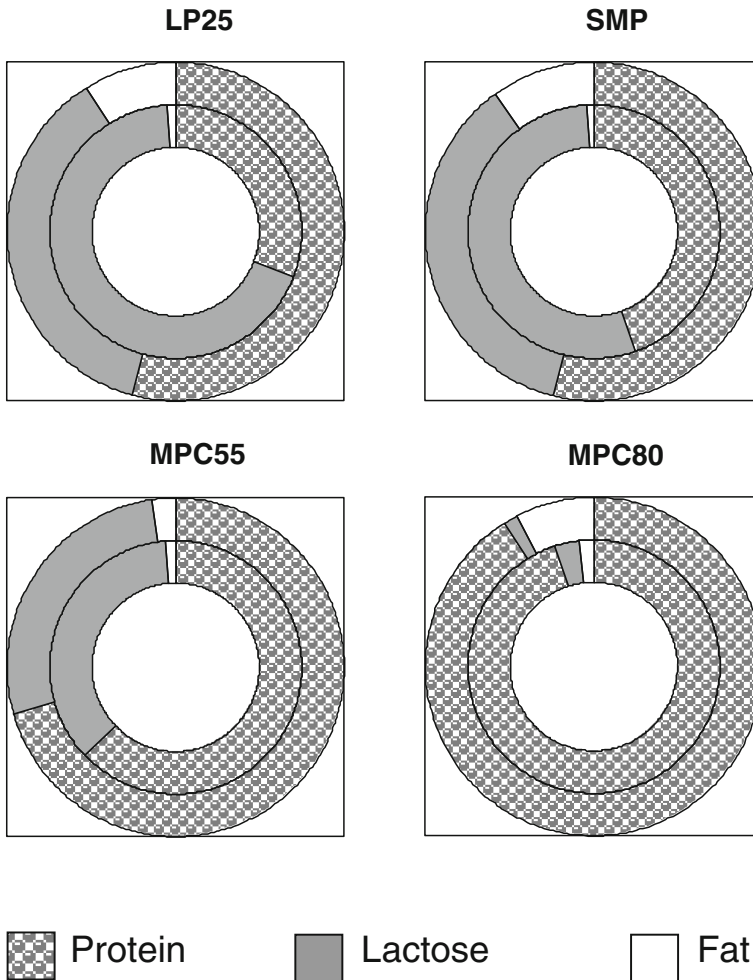
High-fat dairy powders (where fat is the major component, e.g. cream powders with fat content up to 72%) can also be spray-dried. In practice, fouling is a constraint in the manufacture of such products, especially in cyclones and fluid beds. It has been

shown that conditions which initiate an increase in sticking (i.e. a relative humidity above which a major increase in deposition occurs in a particle gun experiment) are not much different for SMP, WMP and cheese powders with up to 55% fat, indicating that the main effect being observed is related to glass transition (Paterson et al. 2005, 2007a, b; Zuo et al. 2007). However, tendency to fouling of high-fat powders in cyclones and upon removal from a spray dryer is thought to be related to the large amounts of fat which are liquid at spray dryer outlet temperatures. It appears therefore that fat causes powder handling problems only when present in amounts large enough to inhibit encapsulation by proteins (Kim et al. 2005). In fat-containing formulations that are rich in sugars, another state transition, namely crystallisation of the sugar, affects the stability of the system during storage by puncturing fat globules and causing the release of free fat (Fäldt and Bergenståhl 1996).

As stickiness specifically involves adhesion at particle surfaces, it is relevant to study surface composition, morphology and electrical charges to understand the influence of surface characteristics on powder stickiness. This could help explain some anomalies between stickiness and glass transition data as the latter is a bulk material property. A range of surface analytical techniques were reviewed by Murrieta-Pazos et al. (2012a). XPS is used to analyse powder particle surface composition. Four non-fat powders varying in protein and lactose contents, which were the subject of previously published studies, were analysed by XPS, which gave composition results for the surface quite different from the bulk composition (Fig. 2). However, even though there were large differences in surface versus bulk composition, the effect of such differences in the protein-to-lactose ratio on surface or bulk  $T_g$  is small compared with the effects of changes in moisture that are encountered near the exit from the drying chamber (Fig. 3). One surprising phenomenon, which is common to other studies, is that even though most milk fat had been removed from those powders, significant amounts of the surface were covered by fat, according to the XPS data (Kim et al. 2002, 2009a; Murrieta-Pazos et al. 2012a, b). This suggests that a very thin layer of fat migrates to the surface during drying due to hydrophobic forces. There is interest in probing a slightly deeper surface layer than the very thin outer layer (~1–10 nm) that is sensed by XPS in order to determine concentration gradients near the surface. Energy-dispersive X-ray analysis techniques with potential to probe deeper are being developed (Murrieta-Pazos et al. 2012b).

#### 4 Application of a physical understanding of stickiness to the spray drying of dairy products

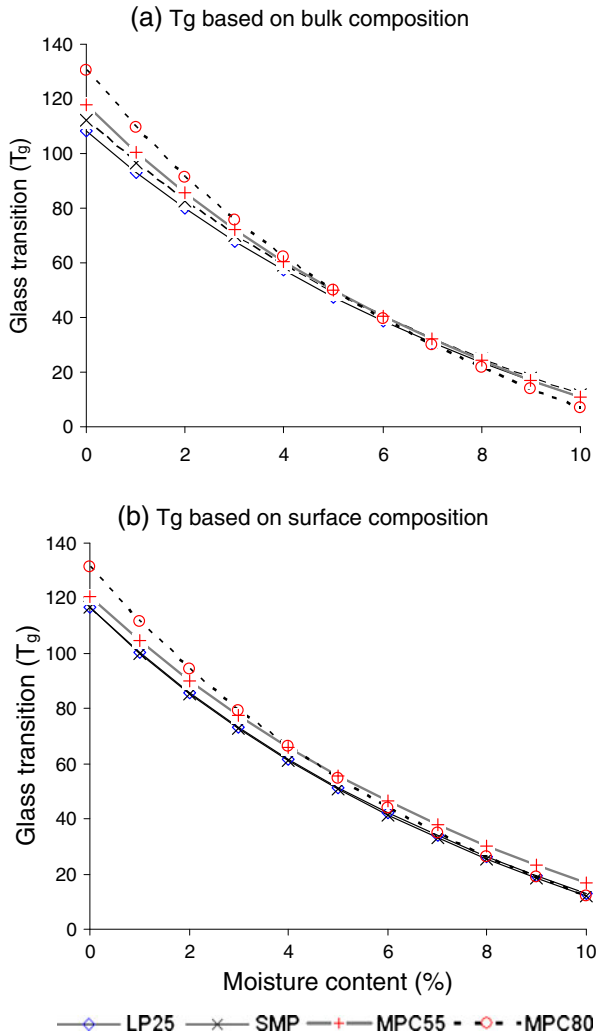
The spray drying chamber is engineered for progressive removal of water, simultaneous with decreasing air temperature and increasing air humidity as the drying air takes up moisture. By combining an understanding of state characteristics (glass transition or melting characteristics, as appropriate) of a food formulation with an understanding of the thermodynamics of drying, it is possible to devise satisfactory manufacturing processes for dried dairy ingredients and formulations. Some approaches involving drying at reduced air temperature have been cited. Other approaches include the use of dehumidified air as low as 60 °C as the inlet air for spray drying (e.g. Bhandari et al. 1997b). Whilst such drying air temperatures can be



**Fig. 2** Doughnut chart showing surface composition by XPS (*outer annulus*) and bulk composition by formulation (*inner annulus*) for four milk protein/lactose powders, with protein concentrations of 25% (*LP25*), 37% (*SMP*), 55% (*MPC55*) and 80% (*MPC80*). The manufacturing procedure for these powders is described in Hogan and O'Callaghan (2010). XPS analysis was carried out at the Materials & Surface Science Institute, University of Limerick

used with vacuum drying (Jaya and Das 2004), they are not commercially feasible for large-scale spray drying. More common approaches include the cooling of chamber walls—with some commercial designs using air brooms that sweep the lower sections of chamber walls with cooled dehumidified air, making powder on or near the wall less sticky by reducing the particle surface temperature as particles approach the wall without increasing the relative humidity (i.e. the water activity level).

The dynamics of fouling and blockage of spray dryers are more complex than glass transition theory alone would predict. Not only do different materials show different rates of adhesion to steel surfaces, but the relative deposition rates (between different products) change over run time; thus, for example, in the early part of a run, maltodextrin had a much lower deposition rate than *SMP*, but after an hour, the rate of



**Fig. 3** Glass transition temperatures based on bulk composition (a) and on surface composition (b) for the four dairy powders referred to in Fig. 2.  $T_g$  was calculated using the Couchman–Karasz equation based on composition data as in Fig. 2 (Couchman and Karasz 1978)

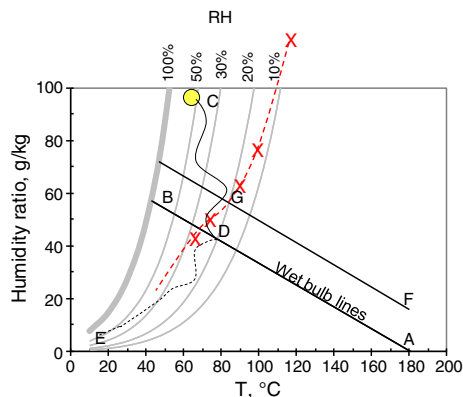
deposition of maltodextrin had exceeded that of SMP (Langrish et al. 2007). Similarly, SMP had greater wall friction but less cohesiveness than WMP or a high-fat powder (Fitzpatrick et al. 2004). New models of sticking with a physical basis are sought which would predict such diverse phenomena.

In general, spray drying can be modelled spatially in one or two dimensions due to geometric symmetry. In the majority of cases, the drying chamber is a vertical tower made up of cylindrical or conical sections and, ideally, the states of powder and air should be symmetrical with respect to the centre line, i.e. axial symmetry should prevail. Sometimes, however, airflow patterns are not axially symmetrical and therefore powder may not be equally dry at the same height on different sides of a chamber (Roustapour et al. 2006). If such asymmetry extends beyond the immediate

atomisation zone, it needs to be addressed as a design issue involving the air distribution system at the point of entry.

A typical locus of drying for a particle undergoing spray drying is superimposed on a psychrometric chart (Fig. 4). The locus  $CD$  depicts the drying of the surface of the particle in some zone as it approaches the exit from the chamber, where it is supposed that surface conditions of the drying particle will approach the surrounding air conditions at a point  $D$  on the “wet bulb line”  $AB$ , the locus of the drying air. A stickiness boundary for the product is drawn on the chart, supposedly corresponding to conditions that would give rise to stickiness at the velocity conditions prevailing at that point. It is of critical importance that the particle at point  $D$  is below the stickiness boundary if it may come in contact with the chamber surface. Upon leaving the chamber, the particle may be subject to further drying and will need to be cooled before storage (point  $E$ ). Just as it is important that point  $D$  be below the stickiness boundary, it is equally important that the cooling/drying locus  $DE$  keeps below the stickiness boundary at all times where the particle may come into contact with equipment surfaces, unless one wishes to promote crystallisation (Yazdanpanah and Langrish 2011a, b). This implies that cooling and air humidity need to be coordinated. It is also obvious that storage conditions require both temperature and humidity control since the product should be stored below its glass transition temperature, which is highly dependent on humidity.

The influence of increasing inlet air humidity can be seen by reference to Fig. 4. An increase in inlet air humidity will alter the locus of drying air to the “wet bulb line”  $FG$ . The air then exits at point  $G$  and the surface of a powder particle will be close to this condition. It is clear that  $G$  is closer than  $D$  to the stickiness boundary. To maintain the same margin against stickiness, it is necessary to reduce the evaporation rate, either by reducing the inlet temperature or increasing the outlet temperature. One way of dealing with this issue is dehumidification of inlet air, which therefore increases the drying capacity.



**Fig. 4** Psychrometric chart showing drying locus for a particle undergoing spray drying in a single-stage dryer. The “wet bulb line”  $AB$  is the locus of the drying air, exiting near point  $D$ . The locus  $CD$  depicts surface conditions for a particle as it approaches the exit. The locus  $DE$  depicts the locus of the particle after leaving the drying chamber as it is cooled and perhaps dried further. The locus  $FG$  is the locus of drying air for an increase in humidity of inlet air. The stickiness zone is indicated by its lower humidity boundary ( $X-X$ )

Thus, where sticking leads to accumulating powder deposits, this tends to occur at points where powder is leaving the drying chamber, where the humid drying air is being separated from the powder or where the powder is being conveyed. Whilst the largest scale spray dryers employ cyclone separators or bag filters with pneumatic conveying to separate the final powder from the drying air, there are some box-type designs of spray dryer, most notably Filtermat and Rogers, which employ mechanical means of conveying the powder using belt-type conveyors or mechanical screw conveyors. In those dryers, whilst cyclones or bag filters are used to remove fine particles, the majority of product falls on the chamber floor and there is a greater tolerance of stickiness issues as powder is removed using mechanical force.

It is believed that particle size plays a role in stickiness, with smaller particles being more prone to stickiness, but not much experimental work has been cited on this (Adhikari et al. 2001).

It has been shown that inlet/outlet drying temperatures and feed solids level influence the surface composition of spray-dried powders (Kim et al. 2009b). Fat on the surface increases as feed solids concentration is reduced. Surface free fat of WMP decreased as inlet/outlet temperatures were raised, but the reason for this is not entirely clear.

## 5 Conclusions

Much scientific progress has been made in understanding the physical phenomena surrounding stickiness. Progress has mainly involved combining the science of soft matter, along with molecular dynamics simulation and/or particle interaction simulation based on physical principles, and various approaches to the measurement of stickiness and related aspects of glass transition. It is anticipated that a more integrated understanding based on molecular dynamics and soft matter physics, along with ever more powerful techniques for analysing particle surfaces, will continue to accelerate the elucidation of this topic.

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