Lactose in dairy systems can exist in various crystalline and non-crystalline forms. These forms affect lactose behaviour, particularly in processing and storage of low-water dairy foods. Crystalline $\alpha$-lactose monohydrate and anhydrous $\beta$-lactose are well-known solid forms of lactose, which are relatively poorly soluble in water. Its occurrence in two anomeric forms, $\alpha$- and $\beta$-lactose, makes its solubility a complex function of temperature. $\alpha$-Lactose has low solubility in water at room temperature, but mutarotation to equilibrium quantities of the $\alpha$- and $\beta$-forms increases the overall solubility of lactose which increases rapidly with increasing temperature, with a more rapid increase in the solubility of $\alpha$-lactose. Liquid dairy systems contain dissolved lactose in a complex chemical environment and lactose is likely to exist in a composition-, temperature- and process-dependent $\alpha/\beta$-ratio. On rapid removal of solvent water from dairy liquids on dehydration or freezing, lactose molecules retain their solution structure and, therefore, amorphous, non-crystalline solid forms of lactose are typical of dairy powders and frozen dairy desserts (Roos, 1995; Hartel, 2001).

Amorphous lactose in dairy solids may often exist in a glassy, solid state or in a syrup-like, super-cooled liquid state. The apparent glass-like solid state results from a very high viscosity exceeding $10^{12}$ Pa s (White and Cakebread, 1966). The state transition of amorphous solid- and liquid-like states occurs over a second-order-type state transition known as the glass transition (White and Cakebread, 1966), as described in Figure 2.1. The glass transition involves no latent heat but it can be observed from changes in heat capacity, thermal expansion coefficient, dielectric properties, various mechanical and flow properties and molecular mobility (White
The glass transition of hydrophilic dairy solids is dominated by that of lactose in which water acts as a softener or ‘plasticizer’ (Jouppila and Roos, 1994a,b). Plasticization by water can be observed as a decrease in the glass transition temperature with increasing water content.

Water plasticization is an important factor contributing to dehydration characteristics and storage stability of dairy solids. A dramatic and well-documented decrease in the stability of dairy powders occurs above a critical water content and corresponding critical water activity (Supplee, 1926; Troy and Sharp, 1930; Herrington, 1934; Lea and White, 1948; King, 1965; Labuza and Saltmarch, 1981; Jouppila et al., 1997; Haque and Roos, 2006). These values of critical water content and water activity correspond to those at which the glass transition of lactose occurs at the storage temperature (Figure 2.2). Exceeding the glass transition conditions of lactose results in dramatic changes in the flow properties of dairy powders and the time-dependent crystallization of lactose (Roos and Karel, 1991c; 1992; Jouppila et al., 1997; Paterson et al., 2005; Haque and Roos, 2004; 2006). Many other physical and chemical changes observed in dehydrated and frozen dairy system have been shown to result from water plasticization and the glass

![Figure 2.1. A schematic presentation of changes in enthalpy, $H$, entropy, $S$, and volume, $V$ around glass transition temperature, $T_g$, and melting temperature, $T_m$. The glassy state is a non-equilibrium state and the glass transition occurs over a temperature range and results in a change of a solid-like material to a syrup-like liquid in sugar systems.](image-url)
The objective of this review is to highlight properties of non-crystalline lactose and its impact on dairy product characteristics at low water contents and in the frozen state. The non-crystalline state of lactose is often a non-equilibrium state showing time-dependent characteristics which may be observed, for example, from changes in flow properties and time-dependent lactose crystallization.

2.1. State Diagram of Lactose

A state diagram may be considered as a ‘map’ which describes conditions at which non-crystalline systems appear as solid glasses or as super-cooled liquids at various water contents and temperatures. State diagrams describe water plasticization behaviour of hydrophilic amorphous solids and the concentration dependence of the glass transition of solutes taking into account ice formation (solvent crystallization) and its effect on solute transition of lactose (Roos and Karel, 1991a; Slade and Levine, 1991; Jouppila et al., 1997; Hartel, 2001).

The objective of this review is to highlight properties of non-crystalline lactose and its impact on dairy product characteristics at low water contents and in the frozen state. The non-crystalline state of lactose is often a non-equilibrium state showing time-dependent characteristics which may be observed, for example, from changes in flow properties and time-dependent lactose crystallization.
concentration at low temperatures. The state diagram of lactose (Figure 2.3) is useful for characterization of the physical state and physical properties of common dehydrated and frozen dairy foods.

State diagrams have been used by Levine and Slade (1988a, 1989) to characterize the effects of frozen storage temperature on food quality which is particularly important to understand the frozen state properties of ice cream and other dairy desserts. State diagrams are available for lactose, milk powders with various fat contents and with hydrolysed lactose (Jouppila and Roos, 1994b; Roos, 2002), lactose protein mixtures (Haque and Roos, 2006) and lactose–salt systems (Omar and Roos, 2006a,b). It appears that lactose governs the solid state of lactose-containing powders but the hydrolysis of lactose results in a significant change to solid properties. This change is a result of the hydrolysis of lactose to glucose and galactose which differ greatly in their sensitivity to water from that of lactose (Jouppila and Roos, 1994a,b). It is also important to note that the glass transition of dairy solids is a property of the hydrophilic, miscible components, often dominated by lactose or its mixtures with added sugar components and

![Figure 2.3. State diagram of lactose.](image-url)

The glass transition temperature ($T_g$) curve at high solids content explains the physical state dependence on temperature and water plasticization. Solvent water crystallization is controlled by equilibrium freezing as defined by solute concentration and kinetically by vitrification at solute concentration of $C_g$ with a glass transition of a maximally freeze-concentrated lactose at $T_g'$ and onset temperature of ice melting at $T_m'$. Goodreads
products of lactose hydrolysis. Water plasticization occurs only in the solids-
non-fat fraction, and state diagrams describe the solids-non-fat properties of
dairy systems.

The lactose–water system is a binary solute–solvent mixture. Water, as
a small molecular mass solvent, acts as a strong plasticizer and a significant
depression of the glass transition temperature, \( T_g \), occurs at low water con-
tents (Slade and Levine, 1991). The plasticization behaviour of amorphous
polymer–solvent systems is often modelled using the Gordon–Taylor rela-
tionship (Gordon and Taylor, 1952), which allows modelling of the glass
transition temperature depression with increasing water content. The Gor-
don–Taylor relationship is shown in equation (1), where \( w_1 \) and \( w_2 \) are weight
fractions of solids and water, respectively, \( T_{g1} \) and \( T_{g2} \) are the glass transition
temperatures of respective components and \( k \) is a constant:

\[
T_g = \frac{w_1 T_{g1} + k w_2 T_{g2}}{w_1 + k w_2}
\]  

(1)

The constant, \( k \), in equation (1) can be derived from experimental data
for \( T_g \) at various water contents (Roos, 1995). Water plasticization of lactose
has been shown to follow this equation which allows its use for establishing
the glass transition curve in the state diagram of lactose (Roos and Karel,
1991a). The Gordon–Taylor equation has also been applied to predict water
plasticization of dairy powders (Jouppila and Roos, 1994b; Haque and Roos,
2006), casein (Kalichevsky et al., 1993a,b) and a number of other food
systems (Roos, 1995). Although numerous values have been reported for
the glass transition temperature of non-crystalline water, the glass transition
temperature for amorphous water is often taken as \(-135^\circ C\) (Sugisaki et al.,
1968). Several equations other than the Gordon–Taylor relationship are
available for predicting the effects of water plasticization and composition
on the \( T_g \) of dairy solids (Roos, 1995).

Most state diagrams show equilibrium melting temperatures of ice at
various water contents and kinetic limitations for ice formation. Ice formation
ceases at temperatures where the equilibrium ice melting temperature
approaches the glass transition of the freeze-concentrated solutes in an unfro-
zen solute matrix. Kinetically limited ice formation may be described as non-
equilibrium ice formation, which is a typical phenomenon in rapidly cooled
carbohydrate solutions and probably the most common form of ice formation
in frozen dairy systems, including ice cream and frozen yoghurt. One of the first
studies reporting non-equilibrium freezing was that of Troy and Sharp (1930),
who found that rapid freezing of ice cream resulted in freeze-concentration and
super-saturation of lactose which, at a sufficiently low temperature, would not crystallize. Several sugars, including lactose, and sugar–protein mixtures form such super-saturated amorphous matrices in frozen systems (Bellows and King, 1973; Roos and Karel, 1991a; Slade and Levine, 1991; Goff et al., 1993; Roos, 1993; Goff, 2002; Singh and Roos, 2005).

State diagrams show the $T_g$ at various water contents. Freezing of water results in separation of ice and concentration of solutes in unfrozen water. Freezing of water ceases as the glassy state of the unfrozen water–solute phase is approached. The glass transition temperature of the maximally freeze-concentrated solute (temperature at which ice formation ceases) with corresponding solute concentration, $C'_g$, onset temperature for ice melting in the maximally freeze-concentrated solution, $T'_m$, equilibrium ice melting temperature, $T_m$ curve, and solubility, are often included in state diagrams. The state diagram of lactose with transition temperatures and corresponding lactose concentrations is shown in Figure 2.3. The most precise $C'_g$ values and corresponding unfrozen water contents, $W'_g$, can be derived from state diagrams established with experimental $T_g$ values (Roos and Karel, 1991b). The solute concentration of maximally freeze-concentrated solute matrices, including that of non-fat milk solids, has been found to be about 80% (w/w), i.e. the unfrozen water content ($W'_g$) is 20% (w/w). These values correspond to solute and water concentrations, respectively, at which ice formation may not occur in freezing, i.e. ice formation is not possible in a system composed of 20% (w/w) water and 80% (w/w) solutes (Roos and Karel, 1991a,b; Roos, 1993; Jouppila and Roos, 1994b). Higher levels of unfrozen water may exist in maximally freeze-concentrated matrices of food polymers such as starch and proteins due to their much higher $T'_g$ values (Roos and Karel, 1991d; Roos, 1995; Singh and Roos, 2005).

### 2.2. Stickiness and Caking

Stickiness and caking are phenomena which may occur when amorphous powder components are plasticized thermally as a result of heating or by exposure to high humidity, resulting in water sorption and plasticization (Peleg, 1977, 1983; Roos, 1995; Lloyd et al., 1996; Paterson et al., 2005; Fitzpatrick et al., 2007). Stickiness and caking of dairy powders are often related to water plasticization of amorphous lactose. Water plasticization may result in glass transition and viscous flow of the non-crystalline lactose at particle surfaces which is observed as stickiness and caking. The surface viscosity of particles is an important property of amorphous powders. Downton et al. (1982) showed that surface viscosity governs the flow properties, stickiness and caking of amorphous powder particles. Levine and Slade (1988b) suggested
that as the viscosity decreased rapidly above the glass transition, amorphous solids could undergo numerous time-dependent structural transformations. These changes in food systems included stickiness and caking of powders, plating of particles on amorphous granules and structural collapse of dehydrated structures.

Williams et al. (1955) found that the viscosity of amorphous glucose above its glass transition was similar to the viscosity of other inorganic and organic glass-forming compounds. Viscosity was related to relaxation times above $T_g$ and followed an empirical relationship known as the William–Landel–Ferry (WLF) equation (2), which was derived from the viscosity data for a number of compounds.

$$\log \frac{\eta}{\eta_s} = \frac{C_1(T - T_s)}{C_2 + (T - T_s)}$$

where $\eta$ is viscosity at temperature, $T$, $\eta_s$ is viscosity at a reference temperature, $T_s$, and $C_1$ and $C_2$ are constants.

The main cause of stickiness is water or thermal plasticization of particle surfaces, which allows a sufficient decrease in surface viscosity and enhances liquid-like behaviour and the development of surface tension for adhesion. Downton et al. (1982) suggested that an increase of temperature or water content caused the formation of an incipient liquid state of a lower viscosity at the particle surface, which resulted in stickiness. Downton et al. (1982) proposed that particles stuck together if sufficient liquid could flow to build strong enough bridges between the particles and that the driving force for the flow was surface tension, which was confirmed for dairy systems by Adhikari et al. (2007).

Stickiness is a time-dependent property. Since viscosity in the glassy state is extremely high, the contact time must be very long to allow adhesion. A dramatic decrease in viscosity above $T_g$ reduces the contact time and causes stickiness which can be related to the time scale of observation. Downton et al. (1982) estimated that a surface viscosity lower than $10^6$–$10^8$ Pa s at a contact time of 1–10 s was sufficient for stickiness. The sticky point was found to decrease with increasing water content. The critical viscosity for stickiness was almost independent of water content, ranging from $0.3 \times 10^7$ to $4.0 \times 10^7$ Pa s, which agreed well with the predicted viscosity range. Wallack and King (1988) reported that the critical viscosity range applied also to other amorphous powders.

Stickiness and caking may also be related to the hygroscopicity of non-crystalline sugars. Brennan et al. (1971) studied stickiness properties of powders in spray drying and they pointed out that two approaches may be
used to reduce the thermoplasticity and hygroscopicity and therefore to solve problems caused by wall deposition in spray drying, i.e. the use of additives as drying aids and the use of specially designed equipment. The sticky point, which describes particle adhesion and stickiness temperature, of amorphous food solids against water content follows an isoviscosity curve with about a constant temperature difference from $T_g$ (Downton et al., 1982; Roos and Karel, 1991a), and the measurement of the sticky point by the method of Lazar et al. (1956) can be considered as a method which, in fact, locates the glass transition of the food solids (Chuy and Labuza, 1994).

Dairy solids-non-fat are plasticized by both temperature and water. Water at a constant temperature may affect the physical properties similarly to temperature at a constant water content. Assuming that the WLF-type temperature dependence applies, the viscosity at a constant water content decreases with increasing temperature. The WLF equation with the ‘universal’ constants $C_1 = -17.44$ and $C_2 = 51.6$, when $T_g$ is the reference temperature (Williams et al., 1955), predicts that an isoviscosity state of $10^7$ Pa s exists at about 20°C above $T_g$, which agrees with the experimental and predicted critical viscosity values for stickiness reported by Downton et al. (1982). The particular importance of the relationship between the sticky point and $T_g$ is that the $T_g$ of amorphous dairy powders can be used as a stability indicator. Thus, knowledge of the $T_g$ and its dependence on water content can be used to evaluate causes of stickiness problems, especially in the production and storage of dairy and other amorphous powders, as described in Figure 2.4.

Caking of sticky powders occurs when sufficient time is allowed for surface contact. According to Peleg (1977), liquid bridging is one of the main inter-particle phenomena which result in caking of food powders. Factors that may cause liquid bridging include water sorption, melting of component compounds (e.g. lipids), chemical reactions that produce liquids (e.g. non-enzymatic browning), excessive liquid ingredients, water released due to crystallization of amorphous sugars and wetting of the powder or equipment. The most common caking mechanism in food powders is plasticization due to water sorption and subsequent inter-particle fusion (Peleg and Mannheim, 1977; Peleg, 1983). Caking of amorphous powders often results from the change of the material from the glassy to a less viscous liquid-like state, which allows liquid flow and the formation of inter-particle liquid bridges. Peleg (1983) pointed out that ‘humidity caking’ is the most common mechanism of caking. Humidity caking is a consequence of an increasing water content, plasticization and depression of $T_g$ to below ambient temperature (e.g. Slade and Levine, 1991). The close relationships between stickiness and glass transition suggest that caking also occurs above the $T_g$ with rates which are defined by the temperature difference, $T-T_g$, which for dairy powders is
highly dependent on solids composition which is particularly important to systems with hydrolysed lactose or modified sugar composition (Jouppila and Roos, 1994a,b; Vega et al., 2005).

2.3. Crystallization and Recrystallization

The non-crystalline state of lactose is a non-equilibrium condition with a high level of super-cooling and a large driving force towards the crystalline, equilibrium state. Lactose crystallization and recrystallization in dairy powders and frozen desserts are glass transition-related, time-dependent phenomena which are governed by the mobility of lactose molecules. Crystallization in the solid, glassy state may not occur as translational mobility of lactose is not possible and crystallization is kinetically limited. Molecules in the glassy state are not able to change their spatial arrangement to the highly ordered, crystalline equilibrium state. At temperatures and water contents exceeding the critical values for the glass transition, molecular mobility increases.

![Glass transition temperature, $T_g$, of skim milk with a schematic representation of liquid droplets during dehydration. Dehydration to a glassy state is required for free-flowing lactose systems. The presence of lower molecular weight sugars would reduce the stickiness zone to lower temperatures and water contents while a shift to higher temperatures can be achieved by mixing lactose with higher molecular weight components. In the wet and semi-dry droplets, lactose exists as a liquid which becomes increasingly viscous as the water content decreases.](image-url)
rapidly and results in lactose crystallization in various forms, depending on
temperature and water content (Haque and Roos, 2005).

Crystallization of amorphous lactose in dairy powders and in ice cream
during storage is one of the principal causes of loss of product quality
(Supplee, 1926; Troy and Sharp, 1930). Supplee (1926) reported that milk
powders sorbed large amounts of water at low storage relative humidities.
This water often induced changes in properties of the powder, and the water
content decreased at higher humidity conditions due to crystallization. Troy
and Sharp (1930) reported that drying of milk and whey by spray drying and
roller drying produced a glass, composed of a non-crystalline mixture of \( \alpha \)-
and \( \beta \)-lactose. Water sorption by whey powders caused plasticization and
subsequent hardening of the material owing to lactose crystallization.

Herrington (1934) found that lactose glasses were stable at room tem-
perature if they were protected from water. The existence of lactose in the
glassy state in dairy products and lactose crystallization at high storage
humidities have been confirmed in numerous studies. These studies have
used polarized light microscopy, electron microscopy, differential scanning
calorimetry (DSC), nuclear magnetic resonance (NMR) and X-ray techni-
ques to analyse the physical state of lactose in dairy powders (King, 1965; Lai
and Schmidt, 1990; Roos and Karel, 1990; Jouppila et al., 1997; Haque and
Roos, 2005). As shown in Figure 2.5, water sorption by most dehydrated
dairy products, which contain lactose, shows a characteristic break in the
sorption isotherm, indicating lactose crystallization (Berlin et al., 1968a,b;
Jouppila and Roos, 1994a,b; Haque and Roos, 2006).

The crystallization behaviour of amorphous lactose in milk products is
also temperature dependent. Berlin et al. (1970) observed that the relative
humidity at which the break in sorption isotherms appeared was dependent
on temperature, which was confirmed by Warburton and Pixton (1978). An
increase in storage temperature shifted the break to a lower relative humidity.
The temperature dependence of the water sorption properties of crystallizing
amorphous sugars can be explained by changes in their physical state. DSC
thermograms of milk powders show a glass transition and a crystallization
exotherm for the amorphous lactose fraction (Jouppila and Roos, 1994b).
Water plasticization decreases the \( T_g \) of lactose and a higher water content
causes lactose crystallization at a lower temperature. Water plasticization of
non-crystalline lactose and associated depression of the \( T_g \) to a lower tem-
perature indicates that the break in the lactose sorption isotherm is both
temperature and time dependent.

Amorphous lactose may crystallize in a complex manner in a number of
crystalline forms and the form produced depends on the relative humidity and
temperature. According to Vuataz (1988), lactose crystallizes as the anhy-
drous \( \beta \)-form at relatively low water activities or as\( \alpha \)-lactose monohydrate
Above a water content of 0.57 at room temperature. At higher temperatures, crystallization behaviour may change according to the stability of the crystalline form at the crystallization temperature. As shown in Figure 2.5, at intermediate water contents recrystallization of $\alpha/C1_{12}$ and $\alpha/C1_{11}/C1_{12}$ mixed forms seems to occur and produce higher amounts of $\alpha/C1_{11}$-lactose monohydrate during storage (Haque and Roos, 2005). Various other components in milk, e.g. proteins and salts, also affect the crystallization properties and the crystalline form produced at different temperature and water conditions (Haque and Roos, 2006; Omar and Roos, 2006a,b).

The kinetics of crystallization at a constant temperature above $T_g$ can be related to water content and water activity, which define the temperature difference, $T - T_g$. Therefore, lactose crystallization may occur above a critical water content or water activity at a constant temperature at a rate defined by the corresponding $T - T_g$ (Roos and Karel, 1992). The rate of lactose crystallization in dairy powders increases also with increasing relative humidity of

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**Figure 2.5.** Sorption isotherm of amorphous lactose. A break in water sorption occurs as a result of lactose crystallization above the critical water content. Crystallization can be observed at varying rates at different storage relative humidities. Recrystallization of anhydrous crystals to $\alpha$-lactose monohydrate crystals may be observed at higher water activities (Haque and Roos, 2005).
storage environment (e.g. Saltmarch and Labuza, 1980; Vuataz, 1988; Jouppila et al., 1997). Increasing relative humidity increases water sorption and water activity, which causes water plasticization and increases the temperature difference, $T - T_g$. The $T - T_g$ of lactose defines the rate of crystallization, as shown in Figure 2.6.

Jouppila and Roos (1994b) determined glass transition temperatures for freeze-dried milk powders, which contained various amounts of fat. The $T_g$ of non-fat solids at various water contents was almost the same as that of lactose (Figure 2.2). The water sorption properties of the non-fat solids were not affected by the fat component. Jouppila and Roos (1994b) developed state diagrams for milk powders, which defined critical values for water content and water activity for stability. Combined $T_g$ and water sorption data suggested that a water content of 7.6 g/100 g of non-fat solids depressed $T_g$ to 24°C. The corresponding water content for pure lactose was 6.8 g/100 g of solids. The critical $a_{w}$ was 0.37. These values, being similar to those shown in Figure 2.2, are in good agreement with several studies which have found critical water contents and storage relative humidities for milk powders based on water sorption properties (e.g. Warburton and Pixton, 1978).

Figure 2.6. Relative nucleation (●) and crystallization (○) rates for lactose at various water activities at room temperature. The glass transition of lactose is defined by water activity, and crystallization occurs above the critical water activity. The rate of nucleation at a low water activity is high but crystal growth occurs slowly which results in a low overall rate of crystallization. The maximum rate and extent of crystallinity (---) is achieved around 0.7 $a_w$ (Jouppila et al., 1997).
Milk powders with lactose hydrolysed to galactose and glucose show no break in their sorption isotherms (San Jose et al., 1977; Jouppila and Roos, 1994a). It was suggested that crystallization of individual sugars in the protein–glucose–galactose mixture was delayed in comparison to lactose crystallization in skim milk and whey powders. Skim milk powders containing hydrolysed lactose showed a $T_g$ well below that of amorphous lactose. Powders produced from skim milk containing galactose and glucose as a result of enzymatic hydrolysis of lactose had an anhydrous $T_g$ at 49°C and a water content of 2.0 g/100 g of solids reduced the $T_g$ to 24°C (Jouppila and Roos, 1994b). Haque and Roos (2006) have shown that the $T_g$ of lactose-containing anhydrous skim milk powders is close to that of lactose at 105°C. However, a number of $T_g$ values for amorphous lactose have been reported, which reflect the sensitivity of the transition to composition and water. Various criteria are also used to locate the transition temperature in DSC thermograms and it may be taken from the onset or mid-point of the transition.

Galactose and glucose show glass transitions at 30 and 31°C (Roos, 1993), respectively. Although Kalichevsky et al. (1993a,b) found that sugars had only a small effect on the $T_g$ of casein, the $T_g$ of milk powders containing hydrolysed lactose seems to be higher than is suggested by the $T_g$ values of the component sugars. The $T_g$ of milk powders is significantly reduced by lactose hydrolysis, which presumably is the main cause of stickiness during processing and storage, as well as of hygroscopic characteristics and higher susceptibility of the powder to non-enzymatic browning reactions. It should also be noted that although lactose is a reducing sugar, the hydrolysis of one mole of lactose produces two moles of reducing sugars, i.e. one mole of galactose and one mole of glucose.

Lactose crystallization in dairy powders results in increasing rates of non-enzymatic browning and other deteriorative changes (Labuza and Saltmarch, 1981; Saltmarch et al., 1981; Miao and Roos, 2004). Saltmarch et al. (1981) found that the rate of browning at 45°C increased rapidly above $a_w$ of 0.33 and showed a maximum between $a_w$ of 0.44 and 0.53. The maximum rate of browning occurred at a lower $a_w$ than was found for other foods. The maximum rate was coincident with extensive lactose crystallization which was observed from scanning electron micrographs. The rate of browning was significantly lower in a whey powder which contained precrystallized lactose. The loss of lysine was also found to be most rapid at water activities which allowed lactose crystallization (Saltmarch et al., 1981). Crystallization of amorphous lactose in closed containers increases water activity very rapidly and accelerates the browning reaction in comparison with the rate of the reaction at the same temperature but at a constant water activity (Kim et al., 1981). Compositional factors and crystallization behaviour of different sugars may also enhance lipid oxidation (Shimada et al., 1991) and browning reactions (Miao and Roos, 2004; Nasirpour et al., 2006).
2.4. Crystallization and Recrystallization in Frozen Systems

The viscosity of a freeze-concentrated solute phase affects time-dependent crystallization phenomena, ice formation and material properties. Levine and Slade (1988a) pointed out that the retarding effect of added maltodextrins on ice recrystallization in ice cream is due to the elevation of the glass transition of a maximally freeze-concentrated solute phase, $T_{g}$. At a sufficiently low temperature, the viscosity of a freeze-concentrated solute matrix becomes high enough to retard diffusion and delay ice formation (Roos and Karel, 1991b). Maximum freeze-concentration may occur at temperatures slightly below the onset temperature of ice melting, $T_{m}$, in the maximally freeze-concentrated material (Figure 2.3). Generally, the $T_{g}$ and $T_{m}$ increase with increasing molecular weight of the solute fraction (Slade and Levine, 1991; Roos and Karel, 1991d).

Lactose crystallization in frozen dairy systems may occur above the glass transition temperature of the maximally freeze-concentrated solute matrix, $T_{g}$. Lactose is one of the least soluble sugars and the loss of quality, including a sandy mouthfeel, resulting from lactose crystallization is well known (Troy and Sharp, 1930; White and Cakebread, 1966). The solubility of lactose at 0°C is only about 12 g/100 g of water and it decreases substantially below the freezing temperature of water as a result of freeze-concentration (Nickerson, 1974). The solubility of lactose also decreases in the presence of other sugars, e.g. sucrose (Nickerson and Moore, 1972), which may significantly facilitate lactose crystallization in frozen dairy desserts and ice cream. However, crystallization of freeze-concentrated solutes can be retarded and greatly reduced by the use of sugar blends and syrups and by the addition of polysaccharides (Hartel, 2001).

Both lactose crystallization and recrystallization of ice in frozen desserts can be reduced by the addition of stabilizers which increase the viscosity of the unfrozen, freeze-concentrated solute phase. Singh and Roos (2005) also showed that in blends of polysaccharides, proteins and sugars, the $T_{g}$ was decreased but the $T_{m}$ increased as a result of retarded ice formation. The polysaccharide, protein (including) polysaccharide and protein stabilizers) and sugar composition seem to be the most important factors in formulation of frozen dairy foods with improved stability against solute crystallization and ice recrystallization.

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