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Dissolution of sucrose crystals in the anhydrous sorbitol melt

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Abstract

The dissolution of a sugar (sucrose as a model) with higher melting point was studied in a molten food polyol (sorbitol as a model) with lower melting point, both in anhydrous state. A DSC and optical examination revealed the dissolution of anhydrous sucrose crystals (mp 192 °C) in anhydrous sorbitol (mp 99 °C) liquid melt. The sucrose–sorbitol crystal mixtures at the proportions of 10, 30, 60, 100 and 150 g of sucrose per 100 g of sorbitol were heat scanned in a DSC to above melting endotherm of sorbitol but well below the onset temperature of melting of sucrose at three different temperatures 110, 130 and 150 °C. The heat scanning modes used were with or without isothermal holding. The dissolution of sucrose in the sorbitol liquid melt was manifested by an increase in the glass transition temperature of the melt and corresponding decrease in endothermic melting enthalpy of sucrose. At given experimental conditions, as high as 25 and 85% of sucrose dissolved in the sorbitol melt during 1 h of isothermal holding at 110 and 150 °C, respectively. Optical microscopic observation also clearly showed the reduction in the size of sucrose crystals in sorbitol melt during the isothermal holding at those temperatures.

Keywords: Sorbitol; Sucrose; Glass transition; Melting; Solubility; Excess enthalpy

1. Introduction

Melting refers to the transformation of an organized solid crystalline structure to the disorganized amorphous liquid state. For a pure material there is a specific melting point, though a range of melting temperatures is observed in a dynamic heating or heat scanning. Impurities normally reduce the melting temperature and can also alter the melting enthalpy.[1,2] In the case of common food and pharmaceutical materials, such as sugars, the melting point can be broadened by strong plasticizers such as water or sorbitol.[3,4] However, the reasons behind this reduction are not explained. Bhandari and Hartel[5] reported the broadening of sucrose melting peak by the presence of fructose and glucose. They found that the primary effects of glucose or fructose addition were a decrease in the onset and peak temperatures of the melting endotherm of sucrose, with only a small change in endset temperatures. The melting endotherm of sucrose was broadened in all cases and the melting enthalpies were reduced by about 3 and 8% at 10 and 20% level of fructose and/or glucose, respectively. They postulated that the fructose and glucose melts may act as solvents for sucrose and probably some exothermic heat of solution of sucrose in fructose and glucose melt counterbalanced endothermic heat of melting (endothermic). This resulted in an overall reduction in melting enthalpy. This generated an interest to the present research which investigated if sucrose, in fact, dissolves in an amorphous liquid melt of other low molecular weight plastisizers, such as monosaccharides or lower molecular weight polyols. As a generalization, this theory considers that the anhydrous liquid melt of one hydrophilic material can serve as a solvent for another compatible anhydrous solid material with a higher melting point. In other words, in an anhydrous food or pharmaceutical system, the lower and higher melting point compounds may behave as solvent and solute, if the lower melting point material is in liquid state (above its melting point).

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The objective of the present research was to examine the above hypothesis using a relevant carbohydrate system. As a conceptual experimental work, this research investigated the dissolution of crystalline sucrose (mp 192 °C) in an anhydrous sorbitol (mp 99 °C) above the endset melting temperature of sorbitol, but below the onset melting temperature of sucrose. Sorbitol was chosen since it melts to a liquid state well below the melting temperature of sucrose and is relatively heat stable. Sorbitol belongs to the polyol group. This polyol is widely used in food and pharmaceutical industries as a replacement of sucrose and other sugars due to its low calorific value, stability, plasticizing property, and it does not cause dental carries. Sorbitol also possesses some functional properties, for example, Bowe[6] reported that a sucrose crystallized in the presence of 5% sorbitol provides directly compressible excipients (without any other binding agents) with an open (porous) crystalline matrix, an optimum property for tablet manufacture in pharmaceutical industries. Besides assisting a fundamental understanding on the subject, our research can also have significant implications for many food and pharmaceutical product systems.

2. Experimental

2.1. Sample preparation

Crystalline sucrose (product no. S-9378, Sigma Aldrich, Germany) and sorbitol (product no. S-1876, Sigma Aldrich, Germany) were used. The materials were used without further purification. The crystalline samples were oven (WTB Binder, Tuttlingen, Germany) dried at 60 °C under vacuum (< 6 Pa absolute) for 12 h prior to the DSC analysis.

2.2. Differential scanning calorimetric analysis

A differential calorimeter (Mettler-Toledo AG DSC821^e) was used for melting and determining the glass transition temperature of the samples. The DSC was calibrated using melting temperatures and enthalpies of *n*-pentane, *n*-hexane, mercury, gallium and indium. The hermetic aluminum pans (40 μL) with lids were used in all DSC scanning. In all the analysis, the sample weight in the pan was maintained between 10 and 14 mg. The temperature scanning was undertaken at the heating and cooling rates of 10 and 20 °C/min, respectively. A flow of 50 mL/min of dry N_2 was used to purge the measuring cell and to prevent water condensation. In all the experiments, three replicates were analyzed.

2.3. Sucrose dissolution analysis using DSC

Mixtures of sucrose and sorbitol were scanned to analyze the dissolution of sucrose in sorbitol melt. The proportions of sucrose/sorbitol studied were 10/100, 30/100, 60/100, 100/100 and 150/100. At first the crystalline sorbitol was weighed in the pan and then the crystalline sucrose was weighed on the bed of the sorbitol. No intentional mixing was done. Since the sucrose was in crystalline form with variable sizes, care was taken to exactly weigh the sucrose crystals. In many instances, the weight of the sucrose exceeded the weight required to maintain the proportion, but the differences were $\pm 2\%$. However, for enthalpy calculations the actual weight of sucrose was considered.

The DSC scanning was undertaken in two modes, dynamic and isothermal. Three maximum scanning temperatures were used, 110, 130 and 150 °C. These temperatures situate above the melting endotherm (endset) of sorbitol but at least 35 °C below the onset melting temperature of sucrose. The dynamic scanning was undertaken to compare the increase in glass transition temperature of anhydrous sorbitol melt resulting from dissolution of sucrose during isothermal holding. During the dynamic scanning, the samples were scanned from 50 °C to the experimental temperatures (110 or 130 or 150 °C) at the rate of 10 °C/min and cooled down to -30 °C at the rate of 20 °C/min and heat scanned to 70 °C at the rate of 10 °C/min. In the isothermal hold scanning mode, the samples were scanned dynamically to experimental temperatures (110, 130 or 150 °C) and held isothermally at these temperatures for 1 h. This time was chosen as it was found to be a minimum time required to detect the changes at the lowest heat scanning temperature (110 °C) applied in this study. The isothermal scanning was used to determine the effect of time on the dissolution of sucrose in sorbitol melt and consequently the increase in glass transition temperature of the melt due to the dissolution of sucrose. After the isothermal holding, the samples were cooled to -30 °C at the rate of 20 °C/min and then heated to 202 °C at the rate of 10 °C/min. The scanning to 202 °C was used to determine the amount of crystalline sucrose remaining in the sample pan based on the melting endotherm area of sucrose. The percentage dissolved sucrose was calculated according to the formula, $\%\ \text{dissolved} = 1 - \Delta H/\Delta H_c$, where ΔH is the heat of fusion in J/g and ΔH_c is the heat of fusion for the 100% crystalline sucrose. The samples were weighed at the end of each DSC scanning to monitor if there was any weight loss due to degradation or other reactions.

2.4. Microscopic observation

The purpose of this experiment was to visualize the reducing size of the sucrose crystals due to dissolution in sorbitol melt. An optical microscope (Leica-Leitz, Switzerland) coupled with a camera was used for this observation. The sucrose crystals were sieved to obtain a size range of 710–850 μm. Four crystals of this size range weighed around 3 mg in average. Thirty mg of sorbitol were weighed on the middle of the glass side. Then, four sucrose crystals were put on top of the sorbitol. The slides containing the samples were held at 110 ± 1, 130 ± 1 and 150 ± 2 °C, in a temperature controlled oven (WTB Binder, Tuttlingen, Germany). The slides were removed at 30 min intervals from the oven and the observation was made quickly using the optical microscope (× 5 magnification) and the photos were taken. A replicate slide with the same amount of samples was also put in the oven in parallel for monitoring if there was any weight loss during the experimental time period.

3. Results and discussion

As described in Section 2, the dissolution of sucrose in sorbitol was evaluated by the upward shift in the glass transition of the melt, reduction in the melting endotherm area of sucrose and microscopic observation of continual reduction in the size of the sucrose crystals with the function of time.

3.1. Melting and glass transition properties of pure sucrose and sorbitol

Table 1 shows the melting properties of pure sorbitol and sucrose crystals. The glass transition of the melt of these carbohydrates is also presented. The onset of glass transition and melting enthalpy values for sorbitol were similar to the values reported by Talja and Roos.[7] The effect of isothermal heating at 150 °C for 60 min on the sorbitol was also investigated by analysing its glass transition temperature (Table 1). There was no change observed in the glass transition temperature indicating that sorbitol was stable at those conditions. In addition, there was no change in the weight and colour of the sorbitol while holding in the oven at 130 °C for 8 h and 150 °C for 2 h. Though no other analysis were undertaken, it is unlikely that sorbitol would have been decomposed or polymerized during these experimental conditions. The midpoint glass transition temperature of sucrose melt was 62 °C, which was slightly lower than the value (67 °C) reported by Roos,[2] probably due to thermal degradation of the sucrose during melting. There was some colour change of the melt, though any noticeable change in weight of the pan at the end of DSC scanning was not recorded.

3.2. Effect of temperature on the dissolution of sucrose crystals

The results obtained from heat scanning with isothermal holding for 1 h of sucrose/sorbitol mixtures at three different temperatures 110, 130 and 150 °C are presented in Tables 2–4, respectively. For comparison purpose, the results for the samples which underwent dynamic scanning (without isothermal holding) to the isothermal temperatures are also presented. The increased isothermal holding temperature increased the glass transition temperature of the melt (Fig. 1). The effect was more pronounced at higher levels of sucrose/sorbitol proportions (Tables 2–4). The dynamic scanning (zero isothermal time) also showed the similar trend at scanning temperature of 150 °C, but the effect was relatively low. At the scanning temperatures of 110 and 130 °C without isothermal holding, the Tg values were unchanged or even showed small increments (Tables 3 and 4) at increased levels of sucrose. This was probably due to an uneven mixing of dissolved sucrose in the sorbitol melt due to the short mixing time and higher viscosity of sorbitol melt at lower temperatures. It should be noted that the mixing of dissolved sucrose in sorbitol was likely to occur purely by diffusion as it was a non-agitated system.

The results based on the melting endotherm area of remaining crystalline (undissolved) sucrose clearly indicated that the amount of dissolved sucrose is higher at higher temperatures (Table 5). Examples of ther-

Table 1
Melting property and glass transition temperature of amorphous melts of sorbitol and sucrose (± indicates standard deviation)

	Glass transition property				Melting endotherm			
	T_{onset} (°C)	$T_{midpoint}$ (°C)	T_{endset} (°C)	ΔCp (J/g °C)	T_{onset} (°C)	$T_{midpoint}$ (°C)	T_{endset} (°C)	ΔH (J/g)
Sorbitol	−6.9 ± 0.2	−1.6 ± 0.1	0.2 ± 0.1	1.0 ± 0.03	96.2 ± 0.9	99.4 ± 0.4	104.3 ± 0.6	177.2 ± 5.2
Sorbitol [a]	−7.2 ± 0.4	−1.7 ± 0.2	0.1 ± 0.1	1.0 ± 0.04				
Sucrose	51.7 ± 0.3	62.1 ± 0.6	67.1 ± 0.5	0.7 ± 0.03	188.3 ± 0.5	191.5 ± 0.6	197.1 ± 0.7	127.0 ± 0.1

[a] Sorbitol melt after an isothermal holding at 150 °C for 60 min.

Table 2
Changes in the glass transition parameters of anhydrous melt due to the dissolution of sucrose in the sorbitol melt during heat scanning at 150 °C, with or without isothermal holding for 1 h at 150 °C

Suc/Sorb [a] ratio	Glass transition temperature (Tg)					
	Without isothermal holding			With isothermal holding		
	T_{onset}	$T_{midpoint}$	T_{endset}	T_{onset}	$T_{midpoint}$	T_{endset}
10/100	−7.7 ± .04	−2.3 ± 0.2	−0.3 ± 0.2	−6.2 ± 0.5	−0.9 ± 0.4	1.4 ± 0.5
30/100	−7.0 ± 0.3	−2.2 ± 0.2	−0.3 ± 0.2	−3.2 ± 0.7	1.3 ± 0.4	6.2 ± 0.7
60/100	−5.9 ± 0.6	−1.2 ± 0.1	0.7 ± 0.1	−0.4 ± 0.9	5.4 ± 1.3	12.8 ± 1.3
100/100	−6.0 ± 0.8	−1.2 ± 1.0	1.4 ± 0.8	0.7 ± 1.0	9.3 ± 1.2	20.1 ± 1.3
150/100	−3.5 ± 0.4	0.7 ± 0.1	6.1 ± 0.6	2.5 ± 0.3	14.1 ± 0.3	28.2 ± 2.4

[a] Suc/Sorb = Sucrose/Sorbitol.

Table 3
Changes in the glass transition parameters of anhydrous melt due to the dissolution of sucrose in the sorbitol melt during heat scanning at 130 °C, with or without isothermal holding for 1 h at 130 °C

Suc/Sorb ratio	Glass transition temperature (Tg)					
	Without isothermal holding			With isothermal scanning		
	T_{onset}	$T_{midpoint}$	T_{endset}	T_{onset}	$T_{midpoint}$	T_{endset}
10/100	−7.0 ± 0.3	−2.6 ± 0.3	−0.7 ± 0.3	−3.8 ± 1.0	−1.3 ± 0.2	0.1 ± 0.6
30/100	−7.3 ± 0.4	−1.7 ± 0.1	0.2 ± 0.1	−4.1 ± 0.1	−0.5 ± 0.4	1.9 ± 0.4
60/100	−6.4 ± 0.3	−1.3 ± 0.2	0.6 ± 0.2	−3.0 ± 0.6	0.6 ± 0.2	4.0 ± 0.3
100/100	−6.9 ± 0.5	−1.4 ± 0.1	0.8 ± 0.3	−3.9 ± 0.8	2.4 ± 0.4	6.6 ± 0.6
150/100	−6.7 ± 0.5	−2.3 ± 0.2	−0.1 ± 0.1	−2.9 ± 0.8	4.2 ± 0.3	8.9 ± 0.2

Table 4
Changes in the glass transition parameters of anhydrous melt due to the dissolution of sucrose in the sorbitol melt during heat scanning at 110 °C, with or without isothermal holding for 1 h at 110 °C

Suc/Sorb ratio	Glass transition temperature (Tg)					
	Without isothermal holding			With isothermal holding		
	T_{onset}	$T_{midpoint}$	T_{endset}	T_{onset}	$T_{midpoint}$	T_{endset}
10/100	−6.8 ± 0.2	−2.0 ± 0.01	−0.2 ± 0.1	−5.9 ± 0.3	−2.1 ± 0.3	−0.4 ± 0.5
30/100	−7.6 ± 0.1	−2.5 ± 0.1	0.5 ± 0.2	−7.2 ± 0.3	−2.5 ± 0.2	1.1 ± 0.03
60/100	−7.0 ± 0.4	−1.8 ± 0.2	−0.04 ± 0.3	−6.6 ± 0.5	−1.8 ± 0.2	0.5 ± 0.4
100/100	−6.8 ± 0.4	−1.9 ± 0.03	−0.01 ± 0.02	−6.7 ± 0.1	−1.4 ± 0.02	1.0 ± 0.1
150/100	−6.8 ± 0.5	−2.0 ± 0.1	−0.13 ± 0.3	−5.7 ± 0.9	0.1 ± 0.4	2.7 ± 0.5

mograms obtained in DSC study demonstrating the reduction in the area of melting endotherm due to the increase in temperature is presented in Fig. 2. Around 85% of the sucrose was found dissolved at 150 °C, whereas at 110 °C this value was around 10%. The decreased solubility of sucrose at lower temperatures can be interpreted with these results. The dissolution rate might have also been influenced by the high viscosity and low diffusivity of solute (sucrose) in the non-agitated system at lower temperatures.

3.3. Effect of proportion of sucrose on the degree of dissolution

The effect of increased amount of sucrose in the sucrose/sorbitol mixture on the dissolution has been pre-

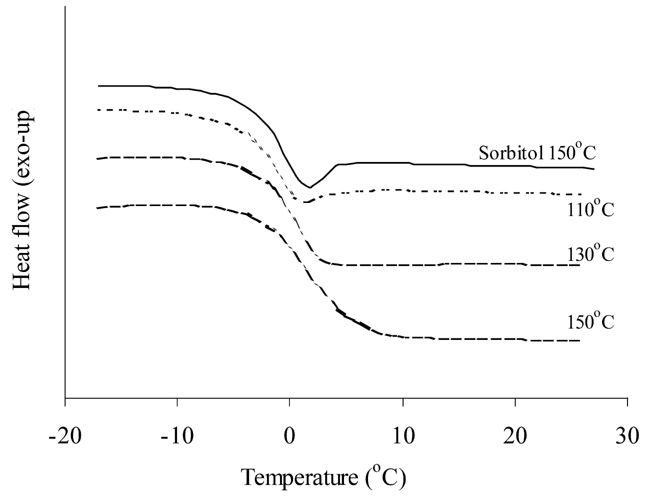

Fig. 1. Representative thermograms indicating the upward shifting of glass transition temperature due to the dissolution of sucrose in the sorbitol (sucrose/sorbitol = 30/100, isothermal holding for 1 h during scanning). Note that the excess enthalpy overshoot is not present in the lower two thermograms (130, 150 °C).

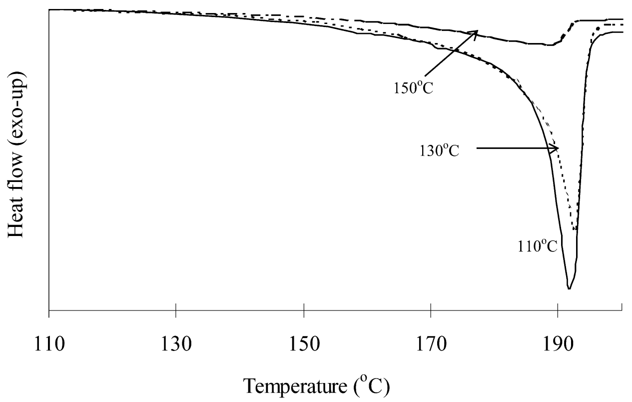

Fig. 2. Representive thermograms illustrating the effect of isothermal holding on the melting endotherm of sucrose for a constant sucrose/sorbitol proportion (100 g sucrose/100 g sorbitol).

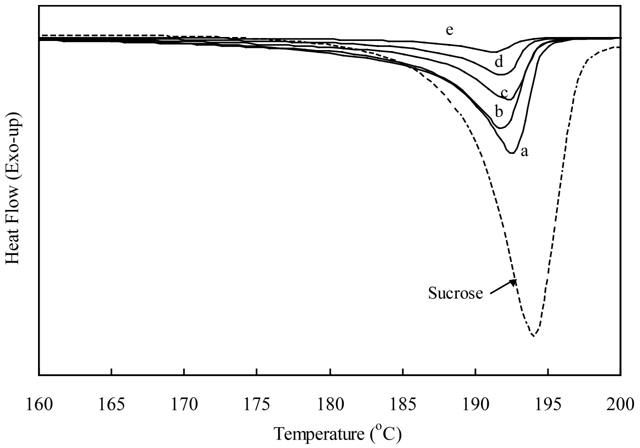

Fig. 3. Representative thermograms indicating the change in the melting endotherms of sucrose due to the increased amount of sucrose in various proportions of sucrose/sorbitol (a = 150/100, b = 100/100, c = 60/100, d = 30/100, e = 10/100).

sented in Table 5. The glass transition temperature of sorbitol melt was increased as the proportion of sucrose increased in the melt. The increase in the glass transition temperature of the melt was a clear indication of dissolution of sucrose (Tables 2–4). The increased dissolution of sucrose had a marked influence on all temperatures given over the glass transition (T_{onset}, T_{midpoint} and T_{endset}). It should be noted that the increased level of sucrose also broadened the transition endotherm (Fig. 3). This could be seen from increased difference between the T_{onset} and T_{endset} as the level of sucrose was increased in the mixture. It is interesting to note that for all levels of sucrose in the mixtures, scanned at 150 °C, the amount of sucrose dissolved was varied narrowly from 80 to 85%, which indicates that the system was still unsaturated with sucrose. Similarly, the variation in the amount of sucrose dissolved at other two temperatures also remained low. In a non-agitated system, the dissolution is governed by the diffusion of sucrose in the liquid sorbitol. It can be presumed that the sucrose is locally dissolved in sorbitol in the vicinity of the crystals. Therefore, the

Table 5
Approximate amount of sucrose dissolved in sorbitol during 1 h isothermal holding at 150 °C

Suc/Sorb ratio	Amount of sucrose crystals dissolved					
	%			g sucrose/100 g sorbitol		
	150 °C	130 °C	110 °C	150 °C	130 °C	110 °C
10/100	84.0 ± 5.6	45.8 ± 3.0	19.2 ± 6.2	8.4 ± 0.6	4.6 ± 0.3	1.9 ± 0.6
30/100	86.3 ± 3.8	29.9 ± 1.7	15.7 ± 1.1	25.9 ± 0.4	9.0 ± 0.2	4.7 ± 0.1
60/100	84.5 ± 2.4	26.6 ± 2.4	16.2 ± 6.4	50.7 ± 1.4	16.0 ± 1.5	9.8 ± 3.9
100/100	80.4 ± 2.9	24.9 ± 2.2	11.0 ± 1.0	80.4 ± 2.9	24.9 ± 2.2	6.6 ± 0.6
150/100	83.3 ± 1.6	28.2 ± 1.5	12.2 ± 0.8	125.0 ± 2.4	28.2 ± 1.5	7.3 ± 0.5

influence of proportion of sucrose/sorbitol did not have strong influence on the rate of dissolution. This is depicted in Fig. 4.

3.4. Effect of dissolution of sucrose on the enthalpy relaxation

An enthalpy recovery endotherm was associated with the glass transition of pure sorbitol (Fig. 1). Such enthalpy relaxation may occur in an amorphous glass below or during glass transition. This relaxation is manifested by appearance of an endotherm or exotherm during the heating cycle in the glass-liquid transition region. In all the experiments, the final cooling temperature and the rate of cooling were kept constant. It was noted that an endothermal enthalpy relaxation decreased in size as the amount of sucrose in the melt increased as a result of sucrose dissolution in liquid sorbitol. Few representative transition curves are presented in Fig. 1. When the level of sucrose was increased, particularly at higher temperatures, no enthalpy relaxation occurred over the glass transition. It can be assumed that the rate of relaxation of amorphous sorbitol molecules interfered by the larger sucrose molecules. It can be clearly observed from Table 6 that the enthalpy relaxation was not noticed above 30 g sucrose/100 g sorbitol, when the mixtures were scanned at 150 °C. This level corresponds to around 26 g of sucrose dissolved in 100 g of sorbitol (Table 5). This suggests that the microstructure is different for mixtures and the relaxation times around the transition are affected by the composition. Shamblin and Zografi[8] found a reduction in the enthalpy relaxation of sucrose due to presence of additives (dextran, trehalose) with higher glass transition temperatures than that of sucrose. They attributed this effect to coupling of molecular motions of mixtures through molecular interactions.

3.5. Optical microscopic observation of dissolution of sucrose crystals

The reducing size of a sucrose crystal due to dissolution in the sorbitol melt at 130 °C is presented in Fig. 5. It took about 2 h for complete disappearance of sucrose at 150 °C. Similarly, it took 7.5 h at 130 °C and more than 48 h at 110 °C. The samples were visually analyzed

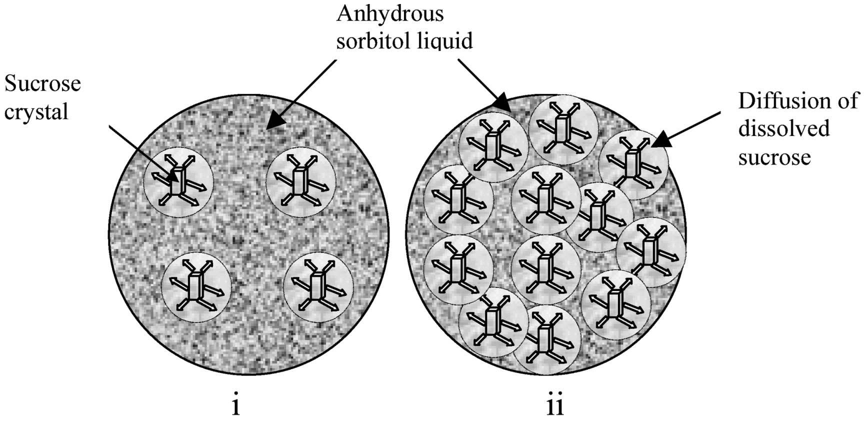

Fig. 4. Schematic representation of diffusion limited dissolution of sucrose crystals in an unagitated anhydrous liquid of sorbitol (i = lower and ii = higher proportion of sucrose/sorbitol). Note that the dissolution is not dependent on the concentration or number of crystals, but on the diffusion around the crystals.

Table 6
Apparent excess enthalpy (J/g) (overshoot above liquidus curve) change due to the dissolution of sucrose in the sorbitol during heat scanning, with or without isothermal holding for 1 h

Suc/Sorb ratio	150 °C		130 °C		110 °C	
	Without isoth.	With isoth.	Without isoth.	With isoth.	Without isoth.	With isoth.
Sorbitol	0.83 ± 0.02	0.89 ± 0.09				
10/100	0.58 ± 0.04	0.24 ± 0.15	0.68 ± 0.04	0.58 ± 0.06	0.85 ± 0.05	0.66 ± 0.07
30/100	0.63 ± 0.12	0	0.74 ± 0.03	0	0.85 ± 0.04	0.61 ± 0.1
60/100	0.29 ± 0.05	0	0.61 ± 0.11	0.12 ± 0.11	0.87 ± 0.14	0.43 ± 0.15
100/100	0	0	0.46 ± 0.13	0.19 ± 0.13	0.86 ± 0.14	0.55 ± 0.11
150/100	0.18 ± 0.04	0	0.51 ± 0.07	0.36 ± 0.08	0.76 ± 0.23	0.63 ± 0.19

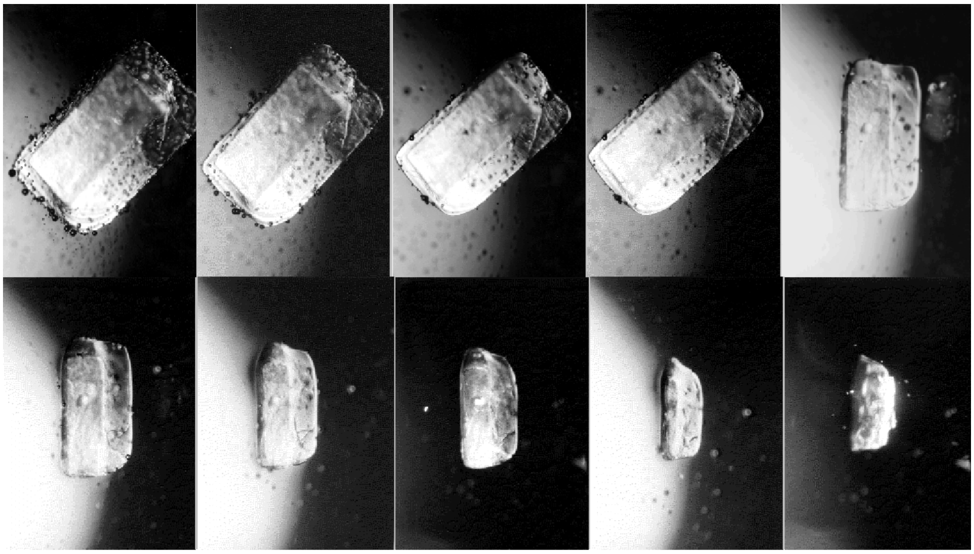

Fig. 5. Optical microscopic photographs (× 5 magnification) of a sucrose crystal dissolving in sorbitol melt (Suc/sorb = 30/100) during isothermal holding at 130 °C. Each picture was taken in every 30 min. Air bubbles trapped during the fusion of sorbitol are seen around the crystals. All photographs were taken in polarized mode, except the last and third last ones were taken in non-polarized light.

for color change and weight was recorded during the isothermal holding periods. At 150 °C, towards the end of the dissolution (1.5 h), there was a slight brown color development around tiny crystals of sucrose. The same observation was made at 130 °C after 5 h of holding at 130 °C. This might have occurred due to degradation of sucrose. However, this probably had only a minor influence on the dissolution because the change in color was observed only when most of the sucrose was dissolved.

4. Conclusion

Dissolution of an anhydrous crystalline sugar in a compatible anhydrous polyol melt was established in the present study. Dissolution of sucrose in sorbitol was confirmed by the increased glass transition of the melt, decreased melting endotherm of sucrose, decreased excess enthalpy of sorbitol in glass transition and microscopic observations. Phase behavior studies of anhydrous sucrose–sorbitol mixtures should be carried out in future studies but in a well agitated (mixed) condition. The results obtained from the present research will have direct implications particularly in food and pharmaceutical systems. Texture, crystallization rate and solubility are some of the properties that can be influenced by the solubilization of one anhydrous compound to another. This research opens a new avenue of investigation for other compatible anhydrous systems.

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