



Why jaggery powder is more stable than solid jaggery blocks

Pankaj Verma^a, Narendra G. Shah^b, Sanjay M. Mahajani^{a,*}



^a Department of Chemical Engineering, Indian Institute of Technology Bombay, Powai, Mumbai, India

^b Centre for Technology Alternatives in Rural Areas, Indian Institute of Technology Bombay, Powai, Mumbai, India

ARTICLE INFO

Keywords:

Non-centrifugal sweetener
Reducing sugars
Crystallinity
Water activity
Moisture sorption isotherm

ABSTRACT

Jaggery, a traditional non-centrifugal sweetener, may be produced in the form of powder or solid blocks. In this study, we compared solid jaggery with powder jaggery on the basis of properties such as crystallinity, crystal size, sorption isotherm and thereby the shelf-life. Powder jaggery shows 7–20 J/g higher heat of melting compared to solid jaggery. A significant difference was observed in the equilibrium moisture content from the sorption isotherm (a_w range 0.11–0.75) of solid and powder jaggery. Intensity count and area under the curve in X-Ray diffraction was higher for powder jaggery samples. From these measurements and analyses, we conclude that powder jaggery was more crystalline and stable, compared to solid jaggery made from the same cane syrup, at all the tested storage conditions.

1. Introduction

Jaggery is a traditional non-centrifugal sweetener popular in many Asian and South American countries. It is generally made in small cottage industries, by concentrating sugarcane juice. Usually, the concentrated syrup is poured into molds to make solid jaggery of different shapes and sizes (Deokate, Bandgar, & Mali, 2009, pp. 1–10; Rao, Das, & Das, 2007; Roy, 1951, pp. 17–43). The jaggery making process involves concentration of clarified sugarcane juice. The concentrated syrup is stirred for a while before being poured into molds for solidification. This stirring facilitates crystallization. Gentle stirring for 10–15 min followed by settling and molding allows formation of conventional solid blocks. On the other hand, to make powder jaggery, the concentrated syrup is stirred continuously and vigorously, and jaggery is not allowed to solidify in bulk.

Jaggery is consumed worldwide and known by different names; Jaggery in most of the Asian and African countries, *Gud* or *Gur* in India, *Kokuto* in Japan, *Naam Taan Oi* in Thailand, *Panela* in Mexico and South America, *Palmzucker* in Germany, *Hukuru* in Sri Lanka, *Muscovado* and *Rapadura* in Brazil and Portugal, etc. India is the largest producer, consumer, and exporter of jaggery meeting close to 60% of the world's demand. In some places, especially in the northern parts of India, production of powder jaggery is also popular. Although powder jaggery fetches more market value nationally and internationally, not many producers are involved in this business, as additional efforts are required for its production. Solid jaggery is easy to make into molds, pack, and transport as compared to its powder counterpart. Due to its

user-friendly nature in handling and utilization in domestic cooking, powder jaggery is gradually gaining popularity among consumers and traders. On the other hand, solid jaggery lumps are difficult to cut in small pieces and require more effort, while powder jaggery is easier to handle and dissolves readily owing to its small particle size and large surface area.

Since powder jaggery is a relatively new product, scant/limited scientific literature regarding its physicochemical properties is available. Rao et al. (2007) and Singh, Singh, Anwar, and Solomon (2011) describe different ways to produce powder jaggery. They have reported the powder jaggery has a lower moisture content (10–30 g/kg) when compared to that of solid jaggery (50–120 g/kg). Their prediction that the shelf-life of powder jaggery would be longer compared to its solid counterpart has not been proven experimentally. Rao, Das, and Das (2010) have studied sorption, glass transition and sticky temperatures of granulated jaggery obtained after milling and reported that the critical moisture content of jaggery typically varies over a range of 10–30 g/kg, above which it is likely to deteriorate faster. The modeling of experimental data for sorption and the glass transition temperature with the help of the Guggenheim, Anderson and de Boer (GAB) model has been tried by many researchers. For solid jaggery, Verma and Narain (1990) explain its sorption behavior with the help of known models. The models are not applicable at high humidities because sugars dissolve in the relatively large amount of the absorbed moisture thereby changing the nature of the solid (Verma & Narain, 1990). Singh, Suman, Arya, Gupta, and Rai (2012) and Chand, Singh, and Kulshrestha (2012) examine the effect of packaging and storage

* Corresponding author.

E-mail addresses: pankajv@iitb.ac.in (P. Verma), nshah@iitb.ac.in (N.G. Shah), sanjaym@iitb.ac.in (S.M. Mahajani).

conditions on the shelf life of jaggery with regards to microbial contamination. These studies reveal that jaggery can be stored for a longer time when under proper storage conditions. Improper packaging material and storage conditions can increase reducing sugars (RS) and moisture, which can result in undesirable changes such as liquefaction, microbial contamination, color and taste changes (Singh et al., 2012). Most of the studies found in the related literature are on solid jaggery, and scant literature is found on powder jaggery. Furthermore, to the best of our knowledge, a comparative product characterization, when made from the same feedstock, is missing in the literature. Data generated in this study indicated that powder jaggery, in terms of its crystallinity and hence the sorption characteristics, differs drastically from the powder made by crushing or milling the solid blocks.

2. Materials and methods

2.1. Chemicals and reagents

Pure sucrose, glucose, and fructose were purchased from Sigma Aldrich, Bangalore, India. Calcium hydroxide was purchased from Loba Chemie, Mumbai, India. Specially dried methanol (moisture < 0.2 ml/l), Karl Fisher reagent and Acetonitrile were purchased from Merck, Mumbai, India. *MagnaFloc®* LT-27 BASF was supplied by Sun Water Technology, Kolhapur, India. Sugarcane (variety CO 86032) was procured from Kolhapur, Mumbai, India.

2.2. Sample preparation

Jaggery was made in the laboratory by the conventional method. The details may be found elsewhere (Ghosh, Shrivastava, & Agnihotri, 1998, pp. 1–15; Roy, 1951, pp. 17–43; Singh et al., 2011). Sugarcane (variety CO 86032) was crushed, and the extracted juice was filtered twice, using a 500 µm filter to remove the large fibers and other suspended impurities. The pH of the juice (2.5 l per batch) was adjusted to 6.5 using freshly prepared milk of lime (saturated solution of calcium hydroxide) to facilitate precipitation of non-sugar impurities. A coagulating agent *MagnaFloc®* LT-27, high molecular weight polyacrylamide, was added (20 mg/l) to the juice which was then heated, and the coagulated impurities were removed. Heating was continued till a viscous syrup was obtained. Heating was stopped when the temperature reached close to 118 °C. Now crystallization of the syrup was done in the following steps.

Step 1: The hot syrup was poured in a flat plate (diameter 0.35 m) and exposed to ambient conditions (typically 23–26 °C and 40–50% RH) for moisture removal and natural cooling.

Step 2: The syrup was then gently stirred for 5–8 min at 20–25 rpm, till a semi-solid mass was produced.

Step 3: A part of this semi-solid mass was poured into a mold for solidification, and thus solid jaggery was prepared.

Step 4: Thereafter the remaining semi-solid mass was spread over a flat plate with a metallic spatula in the form of a thin layer (typically < 1 cm thick). The plate was soon scraped with the help of the same spatula to expose as much syrup as possible to the atmosphere for efficient evaporation.

Step 5: This spreading and scrapping steps were performed, repeatedly- 8–10 times in a very short time interval of 2–3 min. The shear created by this process and the continuous evaporation of free moisture, results in the formation of free-flowing powder jaggery.

Hartel, Ergun, and Vogel (2011) have studied different sweeteners and identified the range of temperature over which crystallization is significant. The steps 4 and 5 are therefore very critical, and the jaggery formation in powdery mass must appear before the temperature drops below 70 °C. If step 5 was not performed as described or if the syrup had more inverted sugars (> 70 g/kg), formed due to inversion, then the

powder with desired crystallinity was not obtained.

Composition and quality of jaggery vary significantly depending on the cane quality, processing method and the operator's skill. The sugar composition in jaggery samples vary typically over a range, 25–100 and 700–900 g/kg for RS (glucose and fructose) and sucrose, respectively. Hence, ten such batches of jaggery were prepared. Each such experiment yielded samples of powder jaggery and solid blocks coming from the same syrup. The pair of these samples was referred as a set; ten such sets were obtained. Three of these ten sets with different RS contents (37, 45, and 65 g/kg) were chosen for further investigations, and analyzed in triplicate, to examine the trends in crystallinity. The samples from the rest of the 7 sets were analyzed only once to confirm the results obtained.

The physicochemical properties of the solid jaggery and powder jaggery samples thus prepared were compared. One set of jaggery samples was also prepared by crushing solid jaggery in a mixer to make a powdered form of jaggery and analyzed along with the other sets, to compare the results.

2.3. Moisture and sugars analysis

The moisture content was determined using a Karl Fischer auto-titration machine (LabIndia, Mumbai, India). Specially dried methanol was used to disperse jaggery samples (~100 mg), and it was titrated against Karl Fisher reagent for the moisture measurement.

Sugars in jaggery (sucrose, glucose, and fructose) were analyzed by High Performance Liquid Chromatography (HPLC) using a zorbax carbohydrate column (4.6 mm × 250 mm) with refractive index detector (both at 40 °C), all purchased from Agilent Technologies, Mumbai, India. The mobile phase was the mixture of acetonitrile and water in a ratio of 75:25 (ml/ml) at a flow rate of 2 ml/min.

2.4. Moisture sorption analysis

For sorption analysis, different salt solutions were used to create environments of varying relative humidity (Greenspan, 1977). Jaggery samples (1–2 g) were spread uniformly over a Petri dish of 4 cm diameter. All the jaggery samples were stored in desiccators in triplicate, and the vacuum was applied to hasten the sorption process. Desiccators were kept in a chamber at a controlled temperature (25, 35 and 45 °C) for about 30 days. The changes in the weight of all the samples were measured to calculate the equilibrium moisture content. After several experiments, it was found that samples stored below $a_w = 0.69$ got equilibrated within 50 h, but the samples stored above $a_w = 0.76$ did not reach equilibrium even after 500 h. Samples stored above $a_w = 0.76$ turned into liquid and developed microbial contamination in a relatively short time, and these samples have not been considered for further analysis.

2.5. Methods used for crystallinity analysis

To check the crystallinity, jaggery samples were analyzed without any pre-treatment using X-ray diffraction (Panalytical, model Empyrean, Germany), Modulated Differential Scanning Calorimetry (TA Instrument, model Q20, New Castle, USA) and Field Emission Gun-Scanning Electron Microscopy (JEOL, model JSM 7600F, Singapore). All the parameters associated with DSC, sorption, and XRD (e.g., scan rate, heating/cooling rate) were standardized by performing several measurements and ensuring the reproducibility of the results for a given sample.

2.5.1. X-ray diffraction (XRD)

The X-ray diffractometer was calibrated using Silicon before the analysis, and samples were analyzed over the diffraction angle (2θ) range of 5–65° (scan rate 0.15°/s) after being uniformly coated onto glass slides. It has been reported that exact quantification of

crystallinity of a complex mixture, like jaggery, is very difficult. The intensity counts and areas under the peak are therefore used as the measure of the extent of crystallinity (Fan & Roos, 2015; Haque & Roos, 2005).

2.5.2. Differential scanning calorimetry (DSC)

Aluminum pans (with pinhole) containing 2–3 mg of samples were used, and the reference pan was kept empty. The temperature program used was as follows: Equilibration at 25 °C, cooling at a rate 10 °C/min to –80 °C, and final heating at 5 °C/min to 250 °C. DSC was calibrated using indium. Heat associated with melting was calculated from the corresponding DSC thermogram using the area under the curve, and it was used as an indication of crystallinity of the samples (Langrish & Wang, 2009; Roos, 1987).

2.5.3. Field emission gun - scanning electron microscopy (FEG-SEM)

The crystal sizes of jaggery samples were determined with the help of FEG-SEM. Jaggery samples were pasted on carbon tape and coated with platinum for the imaging. Images were taken from at least five different spots within a sample and in all, 40–60 crystal particles were considered for the measurement of crystal size using *ImageJ* software.

2.6. Statistical analysis

Results presented are the mean values with their standard deviation of three independent experiments. Analysis of variance (ANOVA) was employed to test the statistical significance ($p < 0.05$). All the calculations were performed using Microsoft Excel (version 2013).

3. Results and discussion

The difference in the stability of powder and solid blocks may be explained by characterizing the products for crystallinity in different ways. Sorption isotherms were used to indicate the stability of the product at different relative humidity. The change in the XRD peak intensity and area at a given diffraction angle (2θ) value, and the change in the heat of melting and glass transition temperature have been used by many researchers as indicators of the extent of crystallinity of the food samples (Fan & Roos, 2015; Miao & Roos, 2005; Roos & Drusch, 2016, chap. 3, pp. 49–77; Wang & Langrish, 2007) and the same approach has been used here to show the significant difference between these two types of products. Other important properties such as wettability, floating tendency, and solubility of powder jaggery and solid jaggery were not significantly different and hence were not studied in detail. All jaggery samples were analyzed for their initial sugars (sucrose, glucose, and fructose) and initial moisture contents before the comparative analysis (Table 1), here SJ stands for solid jaggery and PJ for powder jaggery.

3.1. Moisture sorption isotherm

The equilibrium moisture contents (EMC) of different sets of powder and solid jaggery samples, stored under different humidity and

temperature conditions, have been determined for various samples. Fig. 1 shows the different sorption isotherms of solid and powder jaggery at 25 °C, 35 °C and 45 °C for set 1. Similar results were obtained for set 2 and 3 (Fig. A1 and A2, respectively see Appendix A). Sorption isotherms show that the EMC of powder jaggery was less than that of solid jaggery at any given relative humidity condition. There was an increase in the EMC with an increase in temperature (35 °C and 45 °C) for both jaggery, but even at these temperatures, EMC was higher for solid jaggery samples. The difference in EMC of powder and solid jaggery was much higher for relative humidity (RH) values ranging over 50–80%, compared to lower RH values (10–40%). At RH > 76%, both forms of jaggery turn into liquid-like materials and exhibit similar sorption characteristics. It was observed that the powder form of jaggery prepared by crushing solid jaggery showed similar sorption behavior as solid jaggery (Fig. 2). It may be concluded that the powder made by crushing solid jaggery was characteristically different from the powder jaggery made as per the process described earlier.

It was interesting to note the effect of temperature on the equilibrium moisture content. Fig. 3 shows the isotherm results for the solid jaggery samples at different temperatures. The equilibrium moisture content increases with an increase in temperature at higher humidities (absorption) and reduces with an increase in temperature (desorption) at lower humidities. This causes a reversal in the trend and a crossover in between a_w 0.6–0.7, as seen in Fig. 3. The average value of the triplicate samples was presented on a log scale for better resolution. A similar trend was found in the case of powder jaggery and all the jaggery samples from 10 different sets. A reduction in the equilibrium moisture content (EMC) with an increase in temperature at low water activity is common in foods, and it is explained by the water binding capacity or the increase in the heat of sorption. At a higher water activity, the hygroscopicity of jaggery increases with temperature. Similar trends in the temperature dependency of the moisture sorption isotherm have been reported by many authors. Saravacos, Tsiorvas, and Tsami (1986) and Verma and Narain (1990) have reported it for raisins and jaggery, respectively, while Al-Muhattab, McMinn, and Magee (2002) have mentioned this behavior for various other food products.

The hygroscopicity is not only a function of temperature but also depends on the water activity, the crystal size of the sample and the other components of the food (Mathlouthi & Rogé, 2003). Jaggery typically consists of very small sugar crystals (10–30 µm), thereby increasing hygroscopicity with temperature, especially in an environment with relatively higher humidity (> 70% RH). Koua, Koffi, and Gbaha (2017) and Madiouli, Sghaier, Lecomte, and Sammouda (2012) have attributed this behavior to the opening up of more pores in the cases of other materials.

Since jaggery consists of mostly sucrose with a small fraction of glucose and fructose, sorption isotherm of jaggery was compared with those of pure sucrose, pure glucose, pure fructose and a mixture of all three sugars in a proportion of 94:3:3 (corresponding to the typical jaggery composition on a solid basis). Fig. 4 shows that the moisture sorption of sucrose increases rapidly at around 85% RH, while a more hygroscopic sugar, e.g., fructose starts absorbing significant amounts of moisture from 52% RH. These sorption isotherms are in close

Table 1
Sugars and moisture content (in g/kg) of fresh powder and solid jaggery samples of different sets.

Set Code	Set 1		Set 2		Set 3	
Sample Code	SJ1	PJ1	SJ2	PJ2	SJ3	PJ3
Sucrose	881 ± 34	908 ± 45	843 ± 41	894 ± 53	781 ± 38	824 ± 40
Reducing Sugar	35 ± 3	37 ± 2	45 ± 2	45 ± 3	67 ± 2	65 ± 4
Moisture	84 ± 4*	39 ± 2*	83 ± 3*	46 ± 2*	75 ± 4*	23 ± 2*

SJ1, SJ2, and SJ3: solid jaggery from set 1, set 2, and set 3, respectively; PJ1, PJ2, and PJ3: powder jaggery from set 1, set 2, and set 3, respectively. Results presented are means ± standard deviation ($n = 3$). Asterisks (*) indicate significant differences between the powder and solid jaggery within a given set ($p < 0.05$).

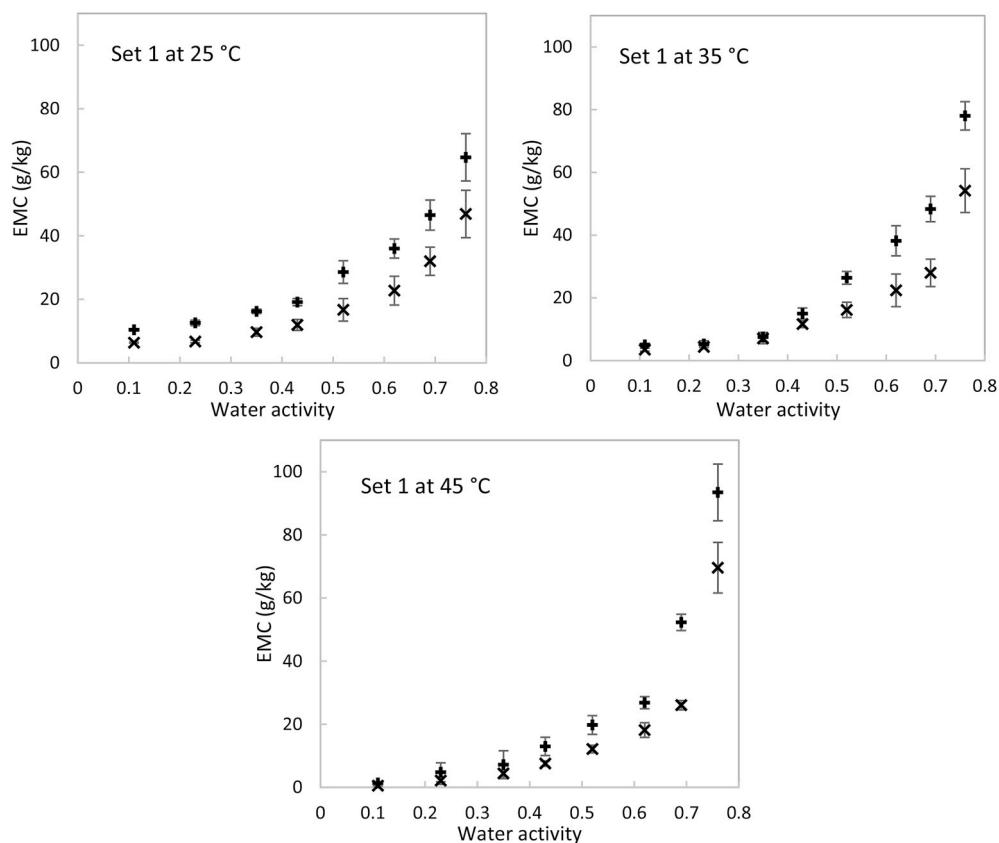


Fig. 1. Sorption isotherms of powder (\times) and solid jaggery (+) samples (set1) at 25 °C, 35 °C and 45 °C, each data shows the mean values of three repetitions with standard deviation, statistical significance difference was proven with $p < 0.05$.

agreement with the literature (Hogekamp, 1997; Moreira, Chenlo, & Torres, 2009; Moreira, Chenlo, Torres, & Prieto, 2017). Jaggery showed a similar trend to the mixture of sucrose, fructose, and glucose in proportion (94:3:3), but moisture sorption isotherm for jaggery increases rapidly from 68% RH, and for the mixture, it started at a slightly higher relative humidity (76% RH) due to its higher degree of crystallinity.

3.2. XRD

Jaggery being a complex mixture of sucrose, glucose, and fructose, exists in the form of a mixture of crystalline and amorphous material, and its XRD pattern consists of many overlapping peaks (Bhandari & Hartel, 2002). A given peak may correspond to any of the three sugars (glucose, fructose, and sucrose). Quantification of the exact value of crystallinity is a difficult task especially in complex mixtures such as foods. Hence, the extent of crystallinity for such systems is usually measured as the difference in the areas under the peak and heights (Fan & Roos, 2015; Haque & Roos, 2005; Jouppila, Kansikas, & Roos, 1998). XRD patterns for sucrose, glucose, and fructose were used as standards (Appendix B Fig. B). We found out few 2θ values where the peak intensity mainly corresponded to a particular sugar, for example, 2θ values of 20.0°, 22.9°, and 25.5° mainly correspond to sucrose, 2θ values of 20.9° and 28.7° correspond to glucose, and 2θ values of 17.2°, 20.5°, 21.0°, and 28.6° mainly correspond to fructose. X-Ray diffraction pattern of a representative set of powder and solid jaggery samples can be seen from Fig. 5. Intensity counts for most of the 2θ values were higher in powder jaggery as compared with solid jaggery. All other samples of powder and solid jaggery were analyzed by XRD, and it was confirmed that powder jaggery shows higher peak intensities compared to solid jagger.

Furthermore, in addition to the intensity, the area under the curve in the XRD patterns which also represents the extent of crystallinity for jaggery samples was calculated at specified 2θ values for comparison.

Powder jaggery samples showed larger areas compared to solid jaggery for most of the 2θ values. The differences in areas and difference in height/intensity count for different 2θ values can be seen from Fig. C1, Appendix C. In most of the cases, the differences, calculated as the count in the powder minus the count in the solid were positive, indicating that the powder jaggery samples are more crystalline compared to solid jaggery.

The set of samples prepared by crushing the solid jaggery was characterized using XRD and compared with its parent solid form. The results showed that for most of the 2θ values, the areas and heights of almost all the peaks are similar (Appendix C, Fig. C2). The time or extent of crushing in the mixer grinder was not vigorous enough to make it amorphous. Crushing the jaggery might have changed the crystal morphology, which has resulted in slight differences in the areas and heights only for certain 2θ values. With the help of DSC analysis and SEM images, we verified that the crushing of solid did not change the crystallinity of the powdered jaggery but did destroy some of the crystal faces. The overall crystallinity of the solid block and the powdered jaggery made by crushing solid blocks was almost similar as the difference in the areas under the peak and the heights (powder – solid) are very small for all the 2θ values, except for 2θ values of 13.3° and 19.9°. This result could be because of the face destruction of the crystals due to crushing. On the contrary, the solid block and jaggery powder made in the process, differ significantly in terms of the crystallinity. The differences (powder – solid) in the areas and heights of almost all the peaks are positive and significant.

3.3. Differential scanning calorimetry (DSC)

The heat associated with the melting of sucrose from the DSC thermogram represents the amount of crystalline sucrose present in the sample. These heat of melting values for solid and powder jaggery

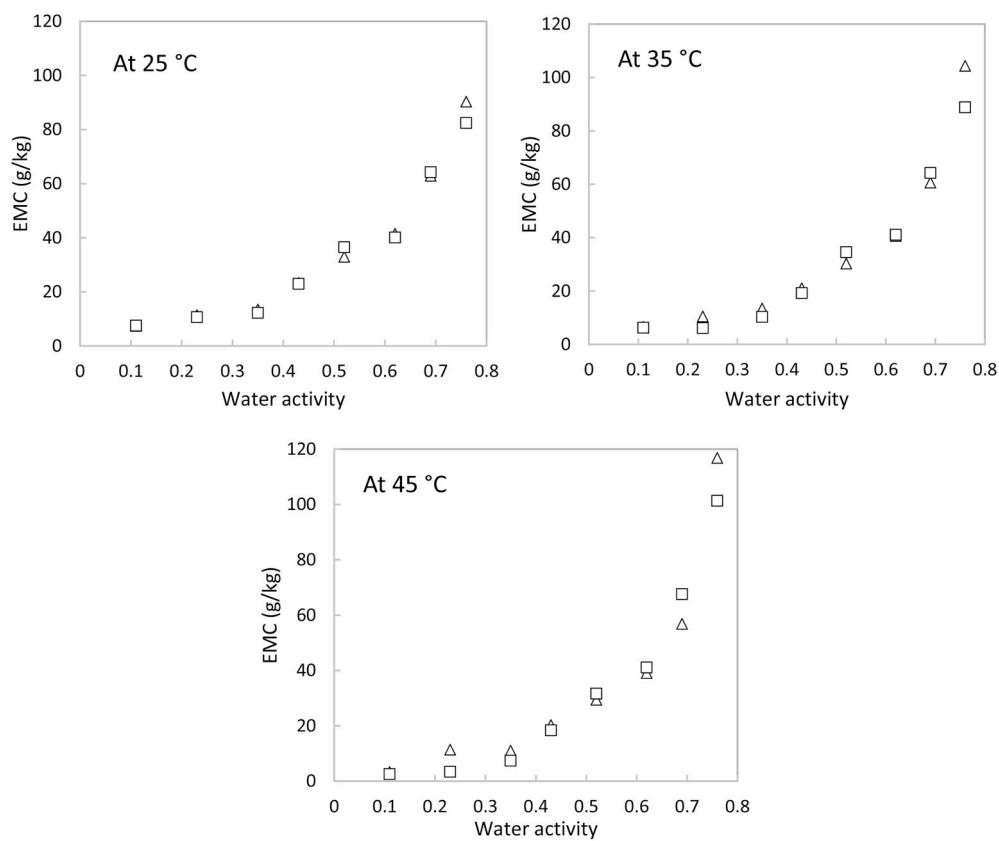


Fig. 2. Comparison of sorption isotherms of solid jaggery (□) and powdered jaggery (Δ) (prepared by crushing the same solid jaggery) at different temperatures. Difference in EMC at different water activity were not statistically significance ($p > 0.05$).

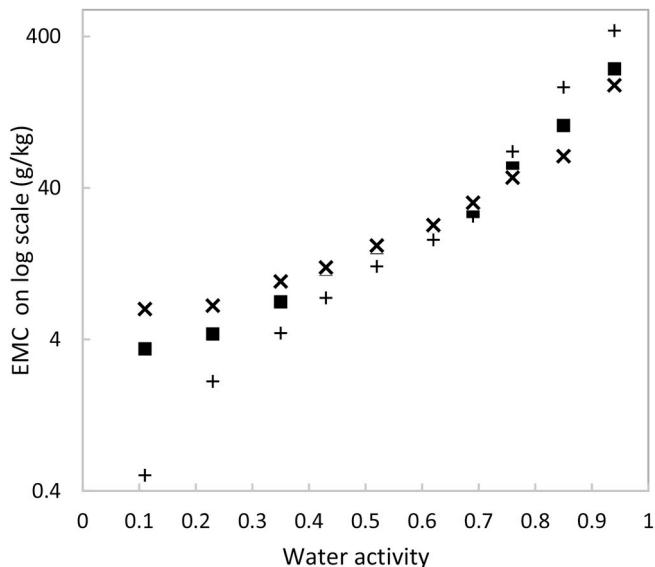


Fig. 3. Sorption isotherm of a jaggery sample (from set 1) at 25 °C (x), 35 °C (■) and 45 °C (+) temperatures (EMCs on a log scale).

products were compared in Table 2. As expected, the values for crystalline sucrose were higher in powder jaggery samples as compared to those in its solid counterpart, in all the sets (Table 2).

3.4. Field emission gun - scanning electron microscopy (FEG-SEM)

FEG-SEM was performed to observe the crystalline structure of jaggery. Jaggery consists of crystals with sizes ranging from 1 to 50 μm .

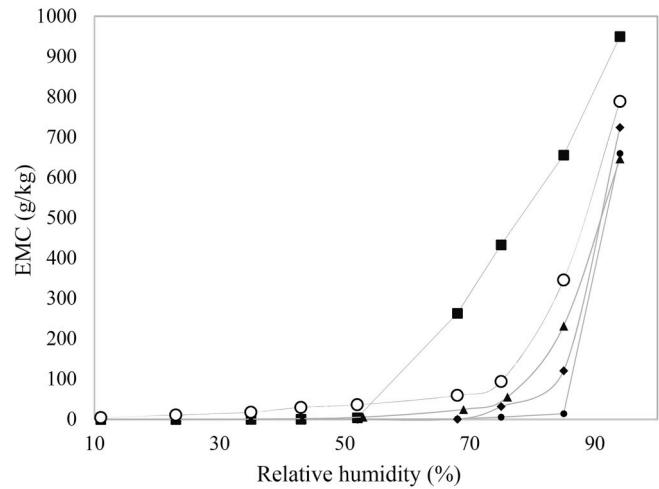


Fig. 4. Comparison of moisture sorption isotherm of jaggery (○) with sucrose (●), glucose (◆), fructose (■), and sugar mixture (▲) (S:G:F: 94:3:3).

(Fig. 6). The sizes of the crystals were measured using *ImageJ* software. SEM images from multiple locations within the sample were taken. The crystals, in general, appear to be coated with the amorphous matter. This amorphous matter was found to be more significant in the solid jaggery samples as compared to powder jaggery (Fig. 6). The crystal size was determined using SEM images. Fig. 7 shows crystal size distributions of two different sets of jaggery samples. The average crystal size of powder jaggery sample was 15 μm ($\sigma = 5$) and that of solid jaggery was 20 μm ($\sigma = 7$) in samples of set 1, while for samples in set 2, they were 12 μm ($\sigma = 6$) and 15 μm ($\sigma = 6$), respectively. The crystal size distribution also confirms that powder jaggery has more small-

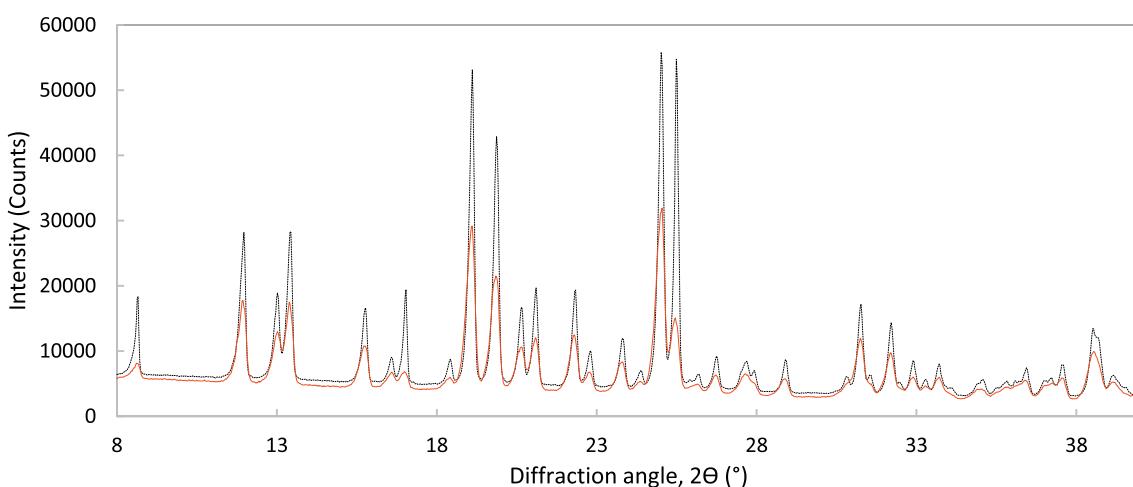


Fig. 5. A comparison of XRD patterns of powder jaggery (---) and solid jaggery (—) from set 1.

Table 2
Heat associated with the melting of sucrose (in J/g) in powder and solid jaggery samples.

Set Code	Set 1		Set 2		Set 3
Sample Code	SJ1	PJ1	SJ2	PJ2	SJ3
Heat	87 ± 2*	104 ± 4*	66 ± 4*	73 ± 1*	67 ± 2*

SJ1, SJ2, and SJ3: solid jaggery from set 1, set 2, and set 3, respectively; PJ1, PJ2, and PJ3: powder jaggery from set 1, set 2, and set 3, respectively. Results presented are means ± standard deviation ($n = 3$). Asterisks (*) indicate significant differences between the powder and solid jaggery within a given set ($p < 0.05$).

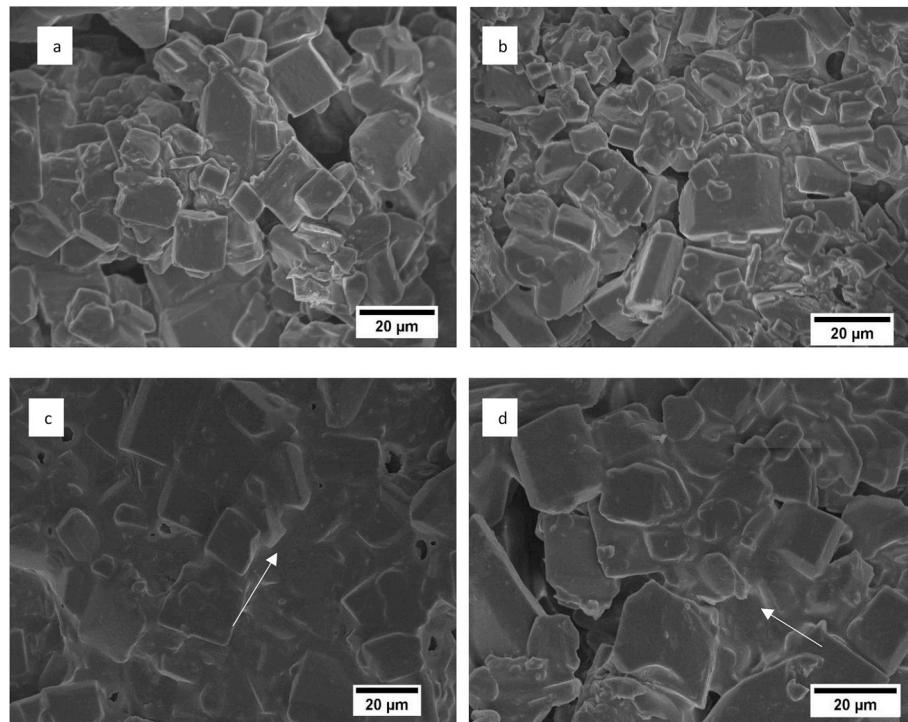


Fig. 6. SEM images, a, b) powder jaggery and c, d) solid jaggery samples, with arrows indicating amorphous matter.

sized crystals than the solid jaggery. This difference could be attributed to the formation of new crystals due to a higher nucleation rate (Bhandari & Hartel, 2002; Liu, Svärd, & Rasmussen, 2015; Ottens & De Jong, 1974), and the breakage of crystals due to shear and stress applied while making the powder jaggery. It should be noted that crystals size in powder jaggery was smaller, but the particle size was still in the range of 100–2500 µm.

3.5. Moisture loss during the crystallization process

The hot syrup ready for crystallization was stirred for about 7–8 min and allowed to solidify. However, powder jaggery was obtained by continuing this process and stirring the semi-solid mass (ready for solidification) for an extended period of 5–10 min. Samples at different time intervals during the stirring process were analyzed using DSC, for

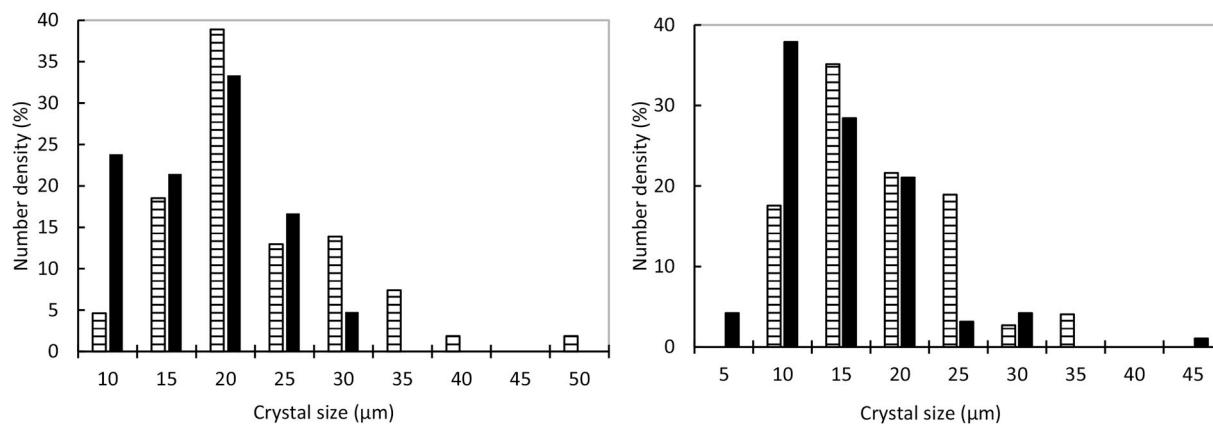


Fig. 7. Crystal size distribution of two different sets of jaggery samples, solid jaggery (□) and powder jaggery (■).

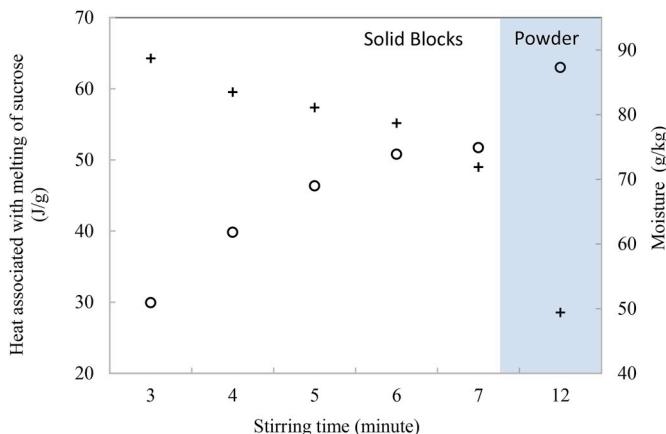


Fig. 8. Change in heat associated with melting (o) and moisture content (+) of a jaggery syrup stirred until the powder is obtained. It may be noted that the time to make solid blocks is slightly over 7 min.

the heat associated with melting, to assess the extent of crystallization and in *Karl Fischer* apparatus for its moisture content. Representative results can be seen from Fig. 8. Samples till the seventh minute are common for both solid blocks and powder. However, the sample at the 12th minute was only for the powder. It was clearly evident that an increase in the heat associated with the melting of sucrose, and a reduction in the moisture content with respect to time, were due to the increased crystallization. This stirring, along with crystallization also resulted in considerable moisture loss. A higher moisture loss in the powder occurs because of more stirring and a large surface area exposed to the atmosphere, and also due to the release of bound water due to increased crystallization (Bhandari & Hartel, 2002). The extra shear applied during the powder-jaggery making process promotes more nucleation, both primary and secondary, which results in the formation of more crystals (Liu et al., 2015; Ottens & De Jong, 1974) and increases the overall crystallinity of jaggery. This makes powder jaggery more stable compared to the solid blocks. Experiments were also performed with multiple sugarcane juice samples, and similar trends were found. We found that this process results in increased crystallization, compared to that in solid jaggery. More crystallization is linked with less water activity and hence better product stability and longer shelf life (Slade & Levine, 1991; Zhou & Roos, 2012).

4. Conclusions

The equilibrium moisture content of powder jaggery was found to be lower, compared to that of the solid jaggery samples. The XRD, DSC and SEM analyses clearly indicate increased crystallinity in the powder

jaggery. The process of making jaggery powder involves prolonged stirring, which leads to more nucleation and growth of sugar crystals. It also results in moisture loss, which is a desirable property from the shelf-life point of view. Even at higher temperature and humidity conditions, powder jaggery was more stable when compared to solid jaggery because of its higher crystallinity.

Acknowledgment

The authors thank the Tata Centre for Technology and Design, IIT Bombay, for financial support. Authors also thank Professor Bhesh Bhandari, from The University of Queensland (Department of Food Processing Technology and Engineering), for his inputs. They also thank Mr. Vishwambhar Patil, Mr. Rohit Kumbhar, Ms. Yamini Jadhav and the project staffs involved in the field trials, for their support in conducting the experiments.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.lwt.2019.04.093>.

References

- Al-Muhtaseb, A. H., McMinn, W. A. M., & Magee, T. R. A. (2002). Moisture sorption isotherm characteristics of food products: A review. *Food and Bioproducts Processing*, 80(2), 118–128. <https://doi.org/10.1205/09603080252938753>.
- Bhandari, B., & Hartel, R. (2002). Co-crystallization of sucrose at high concentration in the presence of glucose and fructose. *Journal of Food Science*, 67(5), 1797–1802. <https://doi.org/10.1111/j.1365-2621.2002.tb08725.x>.
- Chand, K., Singh, A., & Kulshrestha, M. (2012). Jaggery quality effected by hilly climatic conditions. *Indian Journal of Traditional Knowledge*, 11(1), 172–176.
- Deokate, T., Bandgar, D., & Mali, B. (2009). *Marketing and export of jaggery* (1st ed.). Saarbrücken, Germany: Scholars' Press (Chapter 1).
- Fan, F., & Roos, Y. H. (2015). X-ray diffraction analysis of lactose crystallization in freeze-dried lactose-whey protein systems. *Food Research International*, 67, 1–11. <https://doi.org/10.1016/j.foodres.2014.10.023>.
- Ghosh, A. K., Shrivastava, A. K., & Agnihotri, V. P. (1998). *Production Technology of lump sugar- Gur*. Delhi, India: Daya Publishing House (Chapter 1).
- Greenspan, L. (1977). Humidity fixed points of binary saturated aqueous solutions. *Journal of Research of the National Bureau of Standards- A. Physics and Chemistry*, 81A(1), 89. <https://doi.org/10.6028/jres.081A.011>.
- Haque, M. K., & Roos, Y. H. (2005). Crystallization and X-ray diffraction of crystals formed in water-plasticized amorphous spray-dried and freeze-dried lactose/protein mixtures. *Journal of Food Science*, 70(5), E359–E366. <https://doi.org/10.1111/j.1365-2621.2005.tb09977.x>.
- Hartel, R. W., Ergun, R., & Vogel, S. (2011). Phase/state transitions of confectionery sweeteners: Thermodynamic and kinetic aspects. *Comprehensive Reviews in Food Science and Food Safety*, 10(1), 17–32. <https://doi.org/10.1111/j.1541-4337.2010.00136.x>.
- Hogekamp, S. (1997). *About a modified steam jet agglomerator for manufacturing quickly dispersable powders*. Karlsruhe Institute of Technology.
- Jouppila, K., Kansikas, J., & Roos, Y. H. (1998). Crystallization and X-ray diffraction of crystals formed in water-plasticized amorphous lactose. *Biotechnology Progress*, 14(2), 347–350. <https://doi.org/10.1021/bp980010s>.

- Koua, B. K., Koffi, P. M. E., & Gbaha, P. (2017). Evolution of shrinkage, real density, porosity, heat and mass transfer coefficients during indirect solar drying of cocoa beans. *Journal of the Saudi Society of Agricultural Sciences*, 1–11. <https://doi.org/10.1016/j.jssas.2017.01.002>.
- Langrish, T. A. G., & Wang, S. (2009). Crystallization rates for amorphous sucrose and lactose powders from spray drying: A comparison. *Drying Technology*, 27(4), 606–614. <https://doi.org/10.1080/07373930802716391>.
- Liu, J., Svärd, M., & Rasmussen, Å. C. (2015). Influence of agitation on primary nucleation in stirred tank crystallizers. *Crystal Growth & Design*, 15(9), 4177–4184. <https://doi.org/10.1021/cg501791q>.
- Madiouli, J., Sghaier, J., Lecomte, D., & Sammouda, H. (2012). Determination of porosity change from shrinkage curves during drying of food material. *Food and Bioproducts Processing*, 90(1), 43–51. <https://doi.org/10.1016/j.fbp.2010.12.002>.
- Mathlouthi, M., & Rogé, B. (2003). Water vapour sorption isotherms and the caking of food powders. *Food Chemistry*, 82(1), 61–71. [https://doi.org/10.1016/S0308-8146\(02\)00534-4](https://doi.org/10.1016/S0308-8146(02)00534-4).
- Miao, S., & Roos, Y. H. (2005). Crystallization kinetics and X-ray diffraction of crystals formed in amorphous lactose, Trehalose, and lactose/Trehalose mixtures. *Journal of Food Science*, 70(5), E350–E358. <https://doi.org/10.1111/j.1365-2621.2005.tb09976.x>.
- Moreira, R., Chenlo, F., & Torres, M. D. (2009). Simplified algorithm for the prediction of water sorption isotherms of fruits, vegetables and legumes based upon chemical composition. *Journal of Food Engineering*, 94(3–4), 334–343. <https://doi.org/10.1016/j.jfoodeng.2009.03.026>.
- Moreira, R., Chenlo, F., Torres, M. D., & Prieto, D. M. (2017). Statistical criteria for modelling of water desorption isotherms of sugars. Estimation of sucrose hygroscopic properties from glucose and fructose data. *Advances in Food Science and Engineering*, 1(1), 18–27. <https://doi.org/10.22606/afse.2017.11003>.
- Ottens, E. P. K., & De Jong, E. J. (1974). A model for secondary nucleation in a stirred vessel cooling crystallizer. *Kristall und Technik*, 9(8), 873–886. <https://doi.org/10.1002/crat.19740090804>.
- Rao, J. P. V. K., Das, M., & Das, S. K. (2007). Jaggery – a traditional Indian sweetener. *Indian Journal of Traditional Knowledge*, 6(1), 95–102.
- Rao, J. P. V. K., Das, M., & Das, S. K. (2010). Effect of moisture content on glass transition and sticky point temperatures of sugarcane, palmyra-palm and date-palm jaggery granules. *International Journal of Food Science and Technology*, 45(1), 94–104. <https://doi.org/10.1111/j.1365-2621.2009.02108.x>.
- Roos, Y. H. (1987). Effect of moisture on the thermal behavior of strawberries studied using differential scanning Calorimetry. *Journal of Food Science*, 52(1), 146–149.
- Roos, Y. H., & Drusch, S. (2016). *Phase transitions in foods*. Academic press elsevier (2nd ed.). Killington, United Kingdom: Elsevier. <https://doi.org/10.1016/B978-0-12-408086-7.00003-6>.
- Roy, S. (1951). *Monograph on the Gur industry of India*. New Delhi, India: Indian Institute of Sugar Technology (Chapter 2).
- Saravacos, G. D., Tsiorvas, D. A., & Tsami, E. (1986). Effect of temperature on the water adsorption isotherms of sultana raisins. *Journal of Food Science*, 51(2), 381–383. <https://doi.org/10.1111/j.1365-2621.1986.tb11135.x>.
- Singh, J., Singh, R. D., Anwar, S. I., & Solomon, S. (2011). Alternative sweeteners production from sugarcane in India: Lump sugar (jaggery). *Sugar Technology*, 13(4), 366–371. <https://doi.org/10.1007/s12355-011-0110-4>.
- Singh, P., Suman, A., Arya, N., Gupta, R., & Rai, R. K. (2012). Evaluation of sugarcane jaggery shelf life under modified environment: Influence on physico-chemical and microbial properties. *Indian Journal of Sugarcane Technology*, 27(01), 32–36.
- Slade, L., & Levine, H. (1991). Beyond water activity: Recent advances based on an alternative approach to the assessment of food quality and safety. *Critical Reviews in Food Science and Nutrition*, 30(2–3), 115–360. <https://doi.org/10.1080/10408399109527543>.
- Verma, V. K., & Narain, M. (1990). Moisture absorption isotherms of jaggery. *Journal of Stored Products Research*, 26(2), 61–66. [https://doi.org/10.1016/0022-474X\(90\)90001-9](https://doi.org/10.1016/0022-474X(90)90001-9).
- Wang, S., & Langrish, T. (2007). Measurements of the crystallization rates of amorphous sucrose and lactose powders from spray drying. *International Journal of Food Engineering*, 3(4) <https://doi.org/10.2202/1556-3758.1176>.
- Zhou, Y., & Roos, Y. H. (2012). Stability and plasticizing and crystallization effects of vitamins in amorphous sugar systems. *Journal of Agricultural and Food Chemistry*, 60(4), 1075–1083. <https://doi.org/10.1021/jf204168f>.