Original paper

Powder Characteristics of Granulated Sugar in Japan

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The caking of sugar is a major problem in Japan. However, no method has been established to evaluate caking quantitatively. The purpose of this study was to investigate the powder characteristics of granulated sugar in Japan as one of the factors causing caking. The study measured crystal properties, grain size and other powder characteristics of granulated sugar. Samples were collected from four granulated sugar (GS: 500–650 μm) products and 15 fine granulated sugar (FGS: 200–500 μm) products which differed in types of grain sizes. Irregular fine particles were observed in products with a small grain size. GS showed a smaller compression degree and larger Carr index than FGS. As a result of principal component analysis, the products were divided into three groups excluding the particular products.

Keywords: sugar, powder characteristic, grain size, flowability, Carr index, principal component analysis

Introduction

The caking of sugar is a major problem in Japan. Our company has received a yearly average of 157 complaints regarding caking (Figure 1). This could be attributed to Japan having four distinct seasons with extreme climate changes, including changes in humidity and temperature levels (Figure 2). It also seems that customers have become more sensitive to the quality of products. The caking of sugar makes powder more difficult to handle and lowers the manufacturing efficiency. Consequently, investigation of its contributing factors is needed to prevent caking.

In general, the caking of sugar is caused by environmental changes, namely changes in humidity levels, the dissolution and recrystallization of crystal surfaces, and the adhesion of crystals. The crystal goes through four different stages during the caking phenomenon (Rogé and Mathlouthi, 2003).

These four stages are:

A: The pendular stage, at which the sugar is still free flowing;

B: The funicular stage, corresponding to the establishment of permanent contact between grains;

C: The capillary stage, which is reached when the moisture level is high, provoking liquid bridges between grains;

D: The drop stage, obtained when the dissolution of grains is preponderant.

Many complicated factors are related to the caking phenomenon; however, they can be broadly classified into two groups: internal and external factors. The internal factors are powder characteristics, such as grain size, flowability and crystal surface properties, and water activity (or moisture content). External factors are packaging conditions, storage environment (humidity or temperature), storage time, and pressure during storage among other things. Some of these factors explain why caking is a big problem, especially in Japan.

Quantitative evaluation of sugar caking is necessary for assessing countermeasures. However, it is very difficult to establish methods to evaluate the caking phenomenon physicochemically. Based on a previous study (Kanazawa, 1970), establishing a method has the following four main components: (1) to measure powder characteristics related to caking, (2) to adjust the caking of sugar, (3) to measure the physical properties corresponding to the strength of caking, and (4) to evaluate caking quantitatively. However, there are few reports on the powder characteristics of Japanese sugar compared to salt or other food powders (Ito *et al.*, 1999;

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Fig. 1. Complaints about caking (2006-2017)

Fig. 2. Temperature and humidity for Japan (1987-2010)

Takeda, 2013). This study investigated the powder characteristics of granulated sugar in Japan, the first of the four above-mentioned method components.

Materials and Methods

Materials This study used granulated sugar with its simple surface texture. Samples of granulated sugar were collected from 19 products, which differed in types of grain sizes (Table 1):

- Four granulated sugar products (GS: 500-650 μm): three of our products (Mitsui①-③), which were the same products from factories located in different regions of Japan, and a company A product were used.
- Fifteen granulated sugar products (FGS: 200-500 μm): four of our different products (Mitsui①-④), five products from companies A-E, four different products from company F (F①-④) and two different products from company G (G①-②) were used.

Table 1. Studied products

	No.	Product	
GS (500-650 μ m)	1	Mitsui	①
	2		②
	3		③
	4	Company A	
FGS (200-500 μ m)	5	Mitsui	①
	6		②
	7		③
	8		④
	9	Company A	
	10	Company B	
	11	Company C	
	12	Company D	
	13	Company E	
	14	Company F	①
	15		②
	16		③
	17		④
	18	Company G	①
	19		②

Methods

Crystal properties The crystal shape and surface properties were observed under a Miniscope[®] TM3030 (Hitachi High-Technologies, Japan), a low-vacuum electron microscope, and a Digital Microscope KH-7700 (HIROX, Japan).

Grain size The average grain size, grain size distribution and uniformity were measured using a Mastersizer 3000 (Malvern Panalytical, United Kingdom), a laser diffraction scattering type grain size measuring device. The average grain size is expressed in the median diameter, the value of the grain diameter at 50 % in the cumulative distribution (D50). The uniformity, which is one of the indicators of grain size distribution, is calculated by the diameter at 60 % (D60) divided by the diameter at 10 % (D10). In general, when the D60 and D10 are close in value, the powder is not flocculated.

Powder characteristics The repose angle, compression degree, spatula angle and flowability were measured with a Multi tester MT-02 (Seishin Enterprise, Japan). The repose angle is the slope at which the powder will remain in place without sliding. The compression degree expresses the freely settled density (ρ_A) and the tapped bulk density (ρ_T) of the powder, according to the following formula: $(\rho_T - \rho_A) \times 100 / \rho_T$. The spatula angle is the slope angle of the powder, which is measured by inserting a spatula into the powder parallel to the bottom of the container and pulling it out. Carr's flowability index (Carr index) is used as a comprehensive index for the

flowability of a powder. It is calculated from four measured values (repose angle, compression degree, spatula angle, uniformity) and converted to a scale of 25 for each, representing a total of 100. A Carr index greater than 90 is considered to be an index of good flowability, while that below 20 is considered to indicate poor flowability.

Statistical processing The measured values of FGS products were analyzed statistically to clarify the differences among the products. All statistical analyses of the measured values were performed using the Excel statistical software package (BellCurve for Excel; Social Survey Research Information, Japan). The measured values were analyzed using principal component analysis (PCA) in order to group FGS products.

Results

Microphotography images Microphotography images allow the crystal surface properties, which affect powder characteristics, to be understood. There were no clear differences between the four GS products. However, minor differences were observed between our products (Mitsui①-③) and A. Image 1 in Figures 3 and 4 shows Mitsui's standard product. In image 2, the crystal surface of A was smoother and the edges were sharper than Mitsui's product.

Regarding FGS products, irregular crystal shapes were observed in Mitsui① and F③. In addition, more fine particles were seen than in other FGS products such as G① (Figure 5).

Powder characteristics Each of the measured values is presented in Table 2. In a comparison of GS and FGS products, compression degrees tended to be smaller, and Carr indexes of GS tended to be larger compared to FGS. Regarding the FGS products, in particular, it was found that F① and F② products had low spatula angles and large Carr indexes, which showed irregular crystal shapes like Mitsui① and F③ (Figure 6). On the other hand, grain sizes were small and compression degrees were large in the Mitsui① and F③ products with small Carr indexes, showing poor flowability.

Statistical processing Of the 19 samples measured, 15 different FGS products were analyzed to clarify the differences among them. A correlation matrix indicating a relationship is shown in Table 3. The following three relationships were found to have a strong correlation ($p < 0.01$): grain size and compression degree, repose angle and Carr index, and spatula angle and Carr index.

Each measured value was subjected to PCA to acquire first to sixth components. The eigenvalues for the components were 2.468, 2.281, 0.821, 0.332, 0.074 and 0.025. First and second principal components (PC) having large contribution rates were employed (Table 4).

Principal component analysis revealed that the repose angle, spatula angle and Carr index had large principal component load quantities for PC1. On the other hand, the grain size and compression degree had large load quantities for

Fig. 3. Digital microphotography pictures of GS (Mitsui③ and A)

Fig. 4. Microphotography pictures of GS (Mitsui③ and A)

Fig. 5. Microphotography pictures of FGS (Mitsui①, F③ and G①)

PC2, as shown in Table 5.

The principal component scores of each of the products (PC1 vs. PC2) are presented in Figure 7. F① and F② products had large scores in the negative direction of PC1. The other products were divided into the following three groups: positive scores of PC1 and positive scores of PC2, negative scores of PC1 and negative scores of PC2, and positive scores of PC1 and negative scores of PC2.

Discussion

The present study is the first step in the quantitative evaluation of the caking of sugar. To the best of our knowledge, there is no data comparing different granulated sugar products in Japan.

The Carr index is used as an index of the flowability of a powder, which is reduced during caking. Therefore, it was expected that products with small Carr indexes tend to easily cake. However, as a result of our study, no significant

Table 2. Measured values of each product

	No.	Product	Grain size(μm)	Repose angle($^{\circ}$)	Compression degree(%)	Spatula angle($^{\circ}$)	Uniformity	Carr index
GS	1	Mitsui ①	616	32.8	10.1	12.3	1.8	91.5
	2	Mitsui ②	555	32.4	12.2	12.8	1.8	90.0
	3	Mitsui ③	599	31.8	8.4	12.2	2.1	92.0
	4	A	553	32.7	8.5	31.5	2.0	89.0
FGS	5	Mitsui ①	224	33.8	18.9	49.0	2.0	78.0
	6	Mitsui ②	373	30.9	10.5	39.8	1.9	85.0
	7	Mitsui ③	298	28.2	13.5	43.3	1.8	86.0
	8	Mitsui ④	305	33.4	15.6	44.0	2.2	81.5
	9	A	282	34.1	15.4	54.0	1.9	80.0
	10	B	352	26.8	14.1	40.8	1.8	86.0
	11	C	305	31.6	13.1	41.8	1.8	83.0
	12	D	416	36.8	11.4	50.7	1.8	79.0
	13	E	348	38.3	12.1	52.9	1.8	78.0
	14	F①	304	28.6	13.5	12.0	1.7	93.0
	15	F②	282	31.4	17.0	16.4	2.3	88.0
	16	F③	236	31.8	18.4	46.6	2.0	78.0
	17	F④	346	35.0	12.3	51.7	1.6	80.0
	18	G①	308	28.4	14.3	43.4	1.7	86.0
	19	G②	254	33.0	16.5	45.1	1.8	79.5

Fig. 6. Microphotography pictures of FGS (F① and F②)

differences were observed overall. Having said that, the FGS products tended to be smaller than the GS products. It is assumed that when the grain size is small, flowability would be poor because the contact area of the crystal surface is large and inter-grain resistance is greater.

Principal component analysis of FGS aimed to clarify the tendencies of the products. PC1 was explained by repose angle, spatula angle and Carr index, while PC2 was explained by grain size and compression degree. At the same time, the repose angle and spatula angle have a correlation with Carr index. Hence, it seems that PC1 is represented by flowability. However, a comprehensive description of PC1 could not be

found because it comprised positive and negative directions for the principal component load quantities. The reason for this is that the large repose and spatula angles express poor flowability, while, on the other hand, a large Carr index expresses good flowability. In the same way, the grain size would be the most important indicator of PC2, since grain size and compression are strongly correlated. Consequently, these results suggested that flowability and grain size determine the powder characteristics of granulated sugar.

As a result of the principal component scores of each of the products, F① and F② products had different tendencies from the other products, and they had large scores in the negative

Table 3. Correlation matrix of each variable using PCA

	Grain size	Repose angle	Compression degree	Spatula angle	Uniformity	Carr index
Grain size	1.00	0.17	-0.90**	0.10	-0.37	0.11
Repose angle	0.17	1.00	-0.09	0.53	0.07	-0.76**
Compression degree	-0.90**	-0.09	1.00	-0.08	0.56	-0.21
Spatula angle	0.10	0.53	-0.08	1.00	-0.25	-0.86**
Uniformity	-0.37	0.07	0.56	-0.25	1.00	-0.05
Carr index	0.11	-0.76**	-0.21	-0.86**	-0.05	1.00

** $p < 0.01$

Table 4. Eigen value and contribution ratio of each principal component using PCA

	Eigenvalue	Contribution rate	Cumulative contribution rate
PC1	2.47	41%	41%
PC2	2.28	38%	79%
PC3	0.82	14%	93%
PC4	0.33	6%	98%
PC5	0.07	1%	100%
PC6	0.02	0%	100%

Table 5. Principal component load quantities of each measured value using PCA

	Grain size	Repose angle	Compression degree	Spatula angle	Uniformity	Carr index
PC1	0.31	0.82	-0.26	0.89	-0.26	-0.88
PC2	-0.85	0.19	0.93	0.14	0.65	-0.46

direction of PC1. This would be affected by extremely low spatula angles of the products. The other products could be divided into three groups because of their distinctive principal component scores.

Comparison of crystal properties using microphotography images revealed that there were some differences in crystal shape, especially for FGS products. The crystal shapes of Mitsui① and F③ products, with their small grain size and large compression degree, were characterized as asymmetrical particles. These fine particles play the role of binders with crystals and have an adverse influence on flowability. Therefore, crystal properties would be one of the factors in powder characteristics.

However, additional studies are needed to confirm these findings. As a next step, we will further consider the correlation between the measurement of caking intensity and these powder characteristics.

Conclusions

In conclusion, the present study confirmed the following:

- Powder characteristics, such as compression degree and

Carr index, were different between GS and FGS products.

- FGS products with a small grain size and low flowability, e.g., Mitsui① and F③, were observed to have asymmetrical fine particles.
- Flowability and grain size determined the powder characteristics of FGS products, from the results of PCA and the interrelation between measured values.
- It is possible that crystal properties, such as crystal shape and surface properties, affect powder characteristics.
- The correlation between these powder characteristics and caking will be researched.

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Fig. 7. Principal component scores of each product using PCA (axes PC1 and PC2)

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