Powder Technology 272 (2015) 235–240

Contents lists available at ScienceDirect

Powder Technology

journal homepage: www.elsevier.com/locate/powtec

Caking and adhesion free energy of maltitol: Studying of mechanism in adhesion process

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ARTICLE INFO

Article history:
Received 10 May 2014
Received in revised form 24 November 2014
Accepted 5 December 2014
Available online 14 December 2014

Keywords:
Maltitol
Caking
Agglomeration
Adhesion free energy

ABSTRACT

Caking is a common phenomenon of maltitol that causes problems of separation, processing and transportation. To study the mechanism of caking, method of capillary rise was used to measure the contact angles between maltitol particles and relevant liquids. Based on the calculation of adhesion free energy of maltitol particles in different liquids by Lifshitz–van der Waals acid-base theory, the relationship between adhesion free energy and the maltitol particles size has been established. It is concluded that the adhesion free energy of maltitol with rinsing liquid plays a key role in caking. When the adhesion free energy is higher than a critical value (about 60 mJ/m^2), the repulsion between the particles can avoid caking. In general, the adhesion free energy depends on the rinsing liquid used and maltitol particles size. The adhesion free energy increases with increasing the particles size of maltitol and type of liquids (water, ethyl acetate, formamide, ethylene glycol, ethanol, methanol in sequence). Hence, controlling particles size distribution and screening of rinsing liquid are crucial factors to avoid caking.

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1. Introduction

Caking of sugar and sugar polyol is an undesired agglomeration phenomenon during drying process under static, which causes serious problems of separation, processing and transportation. It is therefore of crucial importance to study the mechanism of caking.

Agglomeration is a three-step process: (i) particles collide, (ii) adhere, and (iii) grow together [1]. For caking phenomenon during drying process under static, it is only related to the last two steps. By now, it has been reported that crystal bridges lead to caking of sugar and sugar polyol [2–4]. However, this theory only focused on the growing process and ignored the adhesion process. It has also been reported that adhesion free energy theory, which combined many factors (polarity, interfacial tension, density and viscosity of liquids, particles size and shape) in a value called “adhesion free energy” [1,2,5–7] explained agglomeration of particles in different solvents on energy level, but it has not been used to explain the caking phenomenon under static. Hence, the adhesion process is still lack of studying.

The purpose of this paper is to explain the caking mechanism of maltitol in adhesion process with adhesion free energy theory. In addition, the factors of adhesion free energy were studied to guide rinsing liquid screening for industrial production.

The key to calculate adhesion free energy is contact angle between maltitol particles and liquid, which directly affects the precision of result. Based on the character of maltitol, method of capillary rise was used to measure the contact angles [8]. Though it is not the most accurate method, valuable conclusions are still available through the basic trend.

2. Theory

Adhesion free energy between the particles denotes the affection of repulsion or attraction that can explain the adhesion behavior. It is calculated by Lifshitz–van der Waals acid-base theory [9].

Although it is controversial in the theory about the probe liquids and the values of the polar Lewis acid-base (AB) component, the technique can give important indications, which is the reason that more and more adhesion phenomena have been explained successfully by it [1,5,7]. Therefore, it is worth being used to highlight some indications about caking of sugar and sugar polyol.

The free energy of adhesion between two surfaces (S) of the same chemistry immersed in a liquid (L) is expressed by

$$\Delta G_{SLS} = -2\gamma_{SL} \tag{1}$$

where

$$\gamma_{SL} = \gamma_{SL}^{LW} + \gamma_{SL}^{AB} \tag{2}$$

$$\gamma_{SL}^{LW} = \left(\sqrt{\gamma_S^{LW}} - \sqrt{\gamma_L^{LW}}\right)^2 \tag{3}$$

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http://dx.doi.org/10.1016/j.powtec.2014.12.012
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$$\gamma_{SL}^{AB} = 2\left(\sqrt{\gamma_S^+\gamma_S^-}+\sqrt{\gamma_L^+\gamma_L^-}-\sqrt{\gamma_S^+\gamma_L^-}-\sqrt{\gamma_L^+\gamma_S^-}\right) \tag{4}$$

lead to

$$\Delta G_{SLS} = -2\left[\left(\sqrt{\gamma_S^{LW}}-\sqrt{\gamma_L^{LW}}\right)^2 + 2\left(\sqrt{\gamma_S^+\gamma_S^-}+\sqrt{\gamma_L^+\gamma_L^-}-\sqrt{\gamma_S^+\gamma_L^-}-\sqrt{\gamma_L^+\gamma_S^-}\right)\right] \tag{5}$$

where γ_L is the surface free energy of the liquid, γ_L^{LW} is the nonpolar Lifshitz–van der Waals (LW) component of liquid, the polar Lewis acid-base (AB) components of liquid γ_L^+ and γ_L^- are defined as the non-additive electron-accepting and electron-donating surface free energy parameters, respectively, γ_S^{LW} is the nonpolar Lifshitz–van der Waals (LW) component of solid, γ_S^+ and γ_S^- are the polar Lewis acid-base (AB) components of solid.

For certain liquids the four parameters (γ_L, γ_L^{LW}, γ_L^+, γ_L^-) can be found in the literature. The three unknown surface free energy parameters for the solid (γ_S^{LW}, γ_S^+, γ_S^-) can be estimated by the following equation:

$$(1+\cos\theta)\gamma_L = 2\left(\sqrt{\gamma_S^{LW}\gamma_L^{LW}}+\sqrt{\gamma_S^+\gamma_L^-}+\sqrt{\gamma_S^-\gamma_L^+}\right). \tag{6}$$

By measuring the contact angle for each of three probe liquids, the three unknown parameters of solid are estimated. Then the adhesion free energy can be calculated by Eq. (5).

A positive value of adhesion free energy denotes repulsion between the particles, and a negative value signifies attraction.

As to the probe liquids, water, formamide, and diiodomethane are chosen as a good set [7].

Eq. (6) shows that measuring contact angles between maltitol and liquids is the key to calculate adhesion free energy. Compared with several contact angle measurement techniques [8,10,11], capillary rise was chosen in this paper.

Calculation of contact angle θ is based on Washburn's equation:

$$h^2 = \frac{r\gamma_L\cos\theta}{2\mu}t \tag{7}$$

where h is the height reached by the liquid front at time t, r is the radius of the pores, μ is the viscosity of test liquid.

To overcome difficulties of the visual measurement of the height of advancing liquid front, automatic measurement of the weight gain of penetrating liquid was used for a more accurate result [12].

The relation between the liquid mass (m) and its height in the column is given by:

$$m = \pi R^2 h\rho\varepsilon \tag{8}$$

where R is the inner radius of the column, ρ is the density of test liquid, ε is the porosity of the packed bed.

Substitution in Eq. (7) leads to the modified Washburn's equation as given by:

$$m^2 = \frac{C\rho^2\gamma_L\cos\theta}{\mu}t \tag{9}$$

where

$$C = \frac{r\left(\pi R^2\right)^2\epsilon^2}{2}. \tag{10}$$

C is a material constant which reflects the number and orientation of the capillaries present in the powder sample. C strongly depends on the particle arrangement and packing conditions [10,13]. Hence, uniformed reproducible packing is important for obtaining accurate results [10].

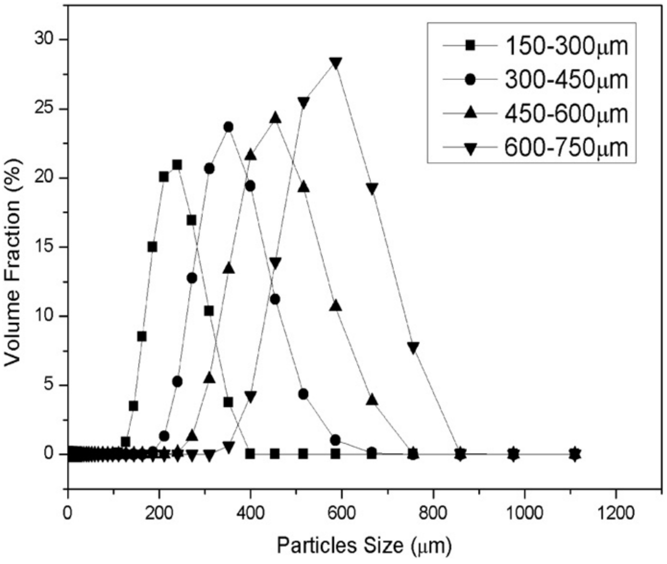

Fig. 1. Size distribution of maltitol in four groups.

3. Experiments

3.1. Materials

Maltitol was chosen as a model because of its strong cohesive properties (purity > 99.9%, purchased from Ningbo Beilunyaxu Chemical Industry Co., Ltd.). Analytical reagent grade of n-hexane, ethanol, formamide, methanol, diiodomethane, ethylene glycol and ethyl acetate were used without further purification. Water was Ultra-Pure.

The maltitol used in the experiments was divided in four groups with quite different particles size by sieves: 150–300 μm, 300–450 μm, 450–600 μm, 600–750 μm, according to Fig. 1.

The shape of maltitol particles is shown in Fig. 2. It has been reported that the difference of caking phenomenon can be observed between flat, cylindrical, and spherical particles [14]. However, discussion about the shape of maltitol is not necessary because of the similar shapes of the whole maltitol samples.

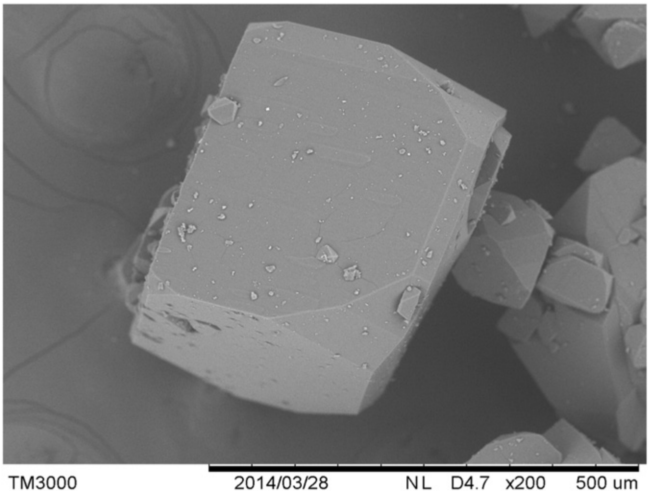

Fig. 2. Shape of maltitol particle obtained by SEM (×200).

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