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Mechanism and inhibition of trisodium phosphate particle caking: Effect of particle shape and solubility

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ABSTRACT

We investigated the influence of particle shape and solubility on the caking behavior of trisodium phosphate by considering the adhesion free energy and crystal bridge theory. Caking of trisodium phosphate during the drying process under static conditions is a two-step process: adhesion followed by crystal bridge formation between particles. The adhesion free energy plays an important role in adhesion. Trisodium phosphate particles cannot adhere to each other and cake when the adhesion free energy is greater than a critical value, which varies with particle shape. Compared with granular particles, cylindrical particles have larger contact area between particles, which results in more crystal bridges forming and a higher caking ratio. Thus, the critical value is about 100 mJ/m^2 for cylindrical particles, but 60 mJ/m^2 for granular particles at 25 °C. Concerning the solubility, when particles are similar shapes and soluble in the rinsing liquid, the caking ratio has a linear relationship with adhesion free energy. However, if the particles are insoluble in the rinsing liquid, caking can be completely prevented regardless of adhesion free energy because no crystal bridges form during the growth process. Hence, caking of trisodium phosphate particles could be inhibited by screening rinsing liquids, and optimizing the particle shape and size distribution.

Introduction

Most solid products in the chemical, pharmaceutical, and food industries are produced as easy to process, free flowing powders. If the products cake, it costs extra time, manpower, and equipment to deal with the caking. Furthermore, 2–10% of product is lost during this process (Thakur, Ahmadian, Sun, & Ooi, 2014; Wahl et al., 2008). It is therefore of great importance to investigate the mechanism of caking and to prepare products without caking.

Some studies have investigated the mechanism of crystal caking during storage. For fine water-soluble crystal particles, partial dissolution and re-crystallization generate crystal bridges that cause particles to consolidate (Bröckel, Wahl, Kirsch, & Feise, 2006; Mauer & Taylor, 2010; Wahl et al., 2008). However, caking of crystals during the drying process under static conditions is a different situation. The products that are separated from the mother solvent usually contain 2–20% liquid content. These wet products are then placed statically to be dried, which is a common situation in industrial production. Compared with storage, particles contain much more liquid during this drying process. As a result, the drying process includes two processes: particles first adhere to each other and then crystal bridges form between particles during the growth process. Crystal bridge theory is therefore not appropriate. Derjaguin–Landau–Verwey–Overbeek (DLVO) theory, which has been used to investigate colloidal particles and biological adhesion (Bramley, Hounslow, Newman, Paterson, & Pogessi, 1997; Clint, 2001), can be used to investigate this caking phenomenon. It involves the balance between attractive van der Waals forces and repulsive electrical double-layer forces (Verwey & Overbeek, 1999). However, the treatment of the short range forces can lead to errors (Bostrom, Williams, & Ninham, 2001), although it has been modified by extra-DLVO forces (Israelachvili, 2011). The adhesion free energy can be used to explain adhesion behavior and agglomeration in different solvents (Ålander & Rasmuson, 2007). In our previous

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Abbreviation: DLVO, Derjaguin–Landau–Verwey–Overbeek.

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Nomenclature

ΔG_{SLS}	adhesion free energy between two surfaces of particles immersed in a liquid (mJ/m^2)
γ^{LW}	nonpolar Lifshitz–van der Waals component (mJ/m^2)
γ^{AB}	polar Lewis acid–base component (mJ/m^2)
γ_S^{LW}	nonpolar Lifshitz–van der Waals components of solid (mJ/m^2)
γ_S^+	nonadditive electron-accepting surface free energy parameter of solid (mJ/m^2)
γ_S^-	electron-donating surface free energy parameter of solid (mJ/m^2)
γ_L^{LW}	nonpolar Lifshitz–van der Waals components of liquid (mJ/m^2)
γ_L^+	nonadditive electron-accepting surface free energy parameter of liquid (mJ/m^2)
γ_L^-	electron-donating surface free energy parameter of liquid (mJ/m^2)
γ_L	surface free energy of liquid (mJ/m^2)
θ	contact angle (°)
m	mass of penetrating liquid (g)
ρ	density of test liquid (g/cm^3)
h	height reached by the liquid front (m)
t	time (s)
r	radius of the pores (m)
μ	viscosity of test liquid (mPa s)
C	material constant
a	caking ratio (%)
m_c	mass of caked trisodium phosphate (g)
M	total mass of trisodium phosphate (g)

work, the caking mechanism in the adhesion process was investigated according to the adhesion free energy. The particle size and the rinsing liquid were found to affect the adhesion free energy, and can be used to avoid adhesion and caking (Chen, Wu, Tang, & Gong, 2015). However, particle shape and solubility should also be investigated to better understand the caking behavior.

In this work, the combined effect of particle shape and solubility on adhesion and growth was investigated. Based on the adhesion free energy and crystal bridge theory, the influences of particle shape and solubility on caking behavior were clarified. Consequently, the caking mechanism of crystals during the dry process under static conditions was determined, and more methods to inhibit caking are summarized.

Theory

The adhesion free energy between particles indicates the affection of repulsion or attraction. A positive value of the free energy of adhesion means repulsion between the particles, whereas a negative value means attraction. The adhesion free energy is calculated by Lifshitz–van der Waals acid–base theory, the Dupré equation, and the Young–Dupré equation (Ålander & Rasmuson, 2007; Van Oss, 2006).

The surface energy is divided into a nonpolar Lifshitz–van der Waals (LW) component γ^{LW} and a polar Lewis acid–base (AB) component γ^{AB}; the latter includes nonadditive electron-accepting and electron-donating surface free energy parameters. For a liquid surface,

$$\gamma_L = \gamma_L^{LW} + \gamma_L^{AB} = \gamma_L^{LW} + 2\sqrt{\gamma_L^+\gamma_L^-}; \tag{1}$$

For a solid surface,

$$\gamma_S = \gamma_S^{LW} + \gamma_S^{AB} = \gamma_S^{LW} + 2\sqrt{\gamma_S^+\gamma_S^-}, \tag{2}$$

where γ_L and γ_S are the surface free energy of the liquid and solid, and γ_L^+, γ_L^-, γ_S^+ and γ_S^- are the nonadditive electron-accepting and electron-donating surface free energy parameters of the liquid and solid.

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

$$G_{SLS} = -2[(\sqrt{\gamma_S^{LW}} - \sqrt{\gamma_L^{LW}})^2 + 2(\sqrt{\gamma_S^+\gamma_S^-} + \sqrt{\gamma_L^+\gamma_L^-} - \sqrt{\gamma_S^+\gamma_L^-} - \sqrt{\gamma_L^+\gamma_S^-})]. \tag{3}$$

For certain liquids, the four parameters (γ_L^+, γ_L^{LW}, γ_L^+ and γ_L^-) can be found in the literature (Della Volpe, Maniglio, Brugnara, Siboni, & Morra, 2004). The Dupré and Young–Dupré equations were used to assess the unknown surface free energy parameters for the solid (γ_S^{LW}, γ_S^+, γ_S^-), Dupré equation can be expressed as

$$\Delta G_{SL} = \gamma_{SL} - \gamma_S - \gamma_L \tag{4}$$

where ΔG_{SL} is the adhesion free energy of a surface immersed in a liquid and γ_{SL} is the surface free energy of the solid/liquid interface. For the solid/liquid interface,

$$\gamma_{SL} = \gamma_{SL}^{LW} + \gamma_{SL}^{AB} \tag{5}$$

where

$$\gamma_{SL}^{LW} = (\sqrt{\gamma_S^{LW}} - \sqrt{\gamma_L^{LW}})^2, \tag{6}$$

$$\gamma_{SL}^{AB} = 2(\sqrt{\gamma_S^+\gamma_S^-} + \sqrt{\gamma_L^+\gamma_L^-} - \sqrt{\gamma_S^+\gamma_L^-} - \sqrt{\gamma_L^+\gamma_S^-}). \tag{7}$$

Combining Eqs. (1), (2), and (4)–(7) with the Young–Dupré equation gives

$$\Delta G_{SL} = -\gamma_L(1 + \cos\theta) \tag{8}$$

Which leads to

$$(1 + \cos\theta)\gamma_L = 2(\sqrt{\gamma_S^{LW}\gamma_L^{LW}} + \sqrt{\gamma_S^+\gamma_L^-} + \sqrt{\gamma_S^-\gamma_L^+}), \tag{9}$$

where θ is the contact angle formed between the liquid and the surface of the solid.

With three test liquids (water, formamide, and diiodomethane) (Della Volpe et al., 2004), the three unknown surface free energy parameters for the solid (γ_S^{LW}, γ_S^+, and γ_S^-), can be estimated by Eq. (9). Compressed discs, thin-layer wicking, and capillary rise can be used to measure the contact angles of the test liquids (Nowak, Combes, Stitt, & Pacek, 2013). Compression of samples into a disc has been attempted (Buckton & Newton, 1986), but the unavailable smooth surface led to an unstable droplet. The particle size of the sample used in this study is outside the measurement range of thin-layer wicking (Nowak et al., 2013; Shang, Flury, Harsh, & Zollars, 2008). Capillary rise was therefore chosen. This method cannot give high accuracy (Bruil & van Aartsen, 1974), but basic conclusions can still be drawn through the trend of the contact angle.

The contact angle is calculated by

$$m^2 = \frac{C\rho^2\gamma_L\cos\theta}{\mu}t \tag{10}$$

which is based on the Washburn's equation (Bruil & van Aartsen, 1974):

$$h^2 = \frac{r\gamma_L\cos\theta}{2\mu}t, \tag{11}$$

where m is the mass of penetrating liquid, ρ is the density of test liquid, h is the height reached by the liquid front at time t, r is the

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