



## Original Research Paper

## Insights into the dissolution mechanisms of detergent agglomerates: An approach to assess dissolution heterogeneity

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## ABSTRACT

In this work, the dissolution mechanisms of detergent agglomerates with different binders were investigated in aqueous solution. The dissolution processes of detergent agglomerates were online monitored by using in situ UV–VIS spectrophotometer and electric conductivity probe. Dissolution profiles were correlated by Weibull model to evaluate the time-dependent dissolution rate coefficient and to classify the type of dissolution rate function  $k_r(t)$ . The Kullback-Leibler information distance  $d_{K-L}$  was proposed to assess the degree of dissolution heterogeneity. The results indicate that the sodium linear alkylbenzene sulfonates (NaLAS) and sodium carbonates ( $\text{Na}_2\text{CO}_3$ ) in detergent agglomerates have different dissolution behaviors, and their dissolution rates are influenced by the type and content of binders. Moreover, detergent agglomerates using semi-solid NaLAS paste or liquid linear alkylbenzene sulfonic acid (HLAS) as binders in granulation processes follow different dissolution mechanisms in water.

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## 1. Introduction

In the past few decades, detergent industry has implemented fundamental changes in process technology [1]. Due to the superior activity towards stain removal, detergent powder still takes a large component of the global market in modern society. Agglomeration is one of the main manufacturing techniques in detergent industry. Different binders used in agglomeration process will lead to the difference in products' physical properties such as particle size distribution, density, particle morphology and so on, which determine the dissolving performance of detergent agglomerates and directly affect the consumer satisfaction. Therefore, the dissolution processes of detergent agglomerates have attracted more and more attention.

Semi-solid sodium linear alkylbenzene sulfonates (NaLAS) paste and liquid linear alkylbenzene sulfonic acid (HLAS) are commonly used as binder in agglomeration. Some reports have demonstrated that granulation mechanism was influenced by the viscosity of binder [2–6]. The granulation processes using highly viscous NaLAS paste and liquid HLAS correspond to the mechanism of coating and layering, wetting and coalescence, respectively. However,

few studies have actually confirmed the effect of binder on dissolution behaviors of detergent agglomerates.

Considering the variation of surface area and shape, the dissolution of particles is more complicated than that of tablets with constant surface area [7]. Even though a variety of theoretical models have been developed, discrepancies between theory and experimental data are still present [8–13]. The alternative approach is based on an empirical function proposed by Weibull [14], which has been successfully applied in almost all types of dissolution profiles [15]:

$$\varphi_t = 1 - \exp[-a \cdot (t - t_0)^b] \quad (1)$$

where  $\varphi_t$  is the accumulated fraction of dissolved material,  $a$  is a scale parameter,  $t_0$  is the lag time, and  $b$  is a shape parameter [16] which characterizes the curves as either typical first-order ( $b = 1$ ), S-shaped ( $b > 1$ ) or exponential with a steeper initial slope ( $b < 1$ ) [15]. The Weibull equation is selected as the dissolution kinetic model since it is associated with various time-dependent phenomena that take place as dissolution proceeds including reduction of the diffusion layer and decrease of the surface area with time, which is intensely beneficial for dissolution process understanding [16,17].

As for the solid dissolution process, homogeneous and heterogeneous dissolution are two widely accepted dissolution mechanisms. The homogeneous case indicates that each molecule has

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the same opportunity to enter solution and the dissolution rate is constant with time (i.e. first-order model) [18]. The heterogeneous case can be defined in such a way that the probability to enter solution is not identical for all molecules in solid state, and then the dissolution rate changes with time [15,18]. The latter case is associated with a variety of phenomena that take place as dissolution proceeds, including reduction of the surface area with time, lack of stirring in the microenvironment of the solid solute, surfaces with irregular boundaries and so on [15,16]. Therefore, the dissolution of detergent agglomerates belongs to the heterogeneous case since the surface area continuously decreases and the dissolution rate drastically depends on the geometrical characteristics of the particles. However, the heterogeneity phenomenon is not completely understood at either experimental or model-based level in the dissolution process of detergent agglomerates.

In this study, NaLAS and sodium carbonates ( $\text{Na}_2\text{CO}_3$ ) were selected as the research objects. A series of detergent agglomerates with different contents of NaLAS were granulated employing two kinds of binders, i.e. semi-solid NaLAS paste and liquid HLAS. To demonstrate the dissolution kinetics of each formula, Weibull model was applied to perform data analyses. The Kullback-Leibler information distance  $d_{K-L}$  [18], which gives a quantitative description of the departure from the homogeneous case, was used to assess the dissolution heterogeneity. Moreover, the correlation between granulation and dissolution mechanism was also discussed, which could be indispensable for proceeding product innovation and process development.

## 2. Theory

### 2.1. Kinetic analysis of the time-dependent dissolution rate

The basic mathematical model for dissolution kinetics was first proposed by Noyes and Whitney [19]:

$$\frac{dc}{dt} = k_c(c_s - c_t) \quad (2)$$

where  $k_c$  is the dissolution rate constant,  $c_s$  is the solubility of solids and  $c_t$  is the concentration of dissolved solids. Obviously, this model belongs to the first-order kinetic equation and the dissolution rate constant  $k_c$  is time-independent. This model can be applied to describe the homogeneous condition.

As mentioned above, a time-dependent rate coefficient  $k_t$  should be conceived to simulate the dissolution process under heterogeneous condition:

$$k_t(t) = k \cdot t^{-h} \quad (3)$$

where  $k$  is a constant with units  $(\text{time})^{h-1}$  and  $h$  is a dimensionless constant [15]. Using Eq. (3) for replacing  $k_c$  in Eq. (2), the kinetic model can be written as:

$$\varphi_t = 1 - \exp\left[-\frac{k}{1-h} \cdot (t - t_0)^{1-h}\right] \quad (4)$$

It is notable that the Weibull function can be derived by replacing  $a = k/(1-h)$  and  $b = 1-h$  in Eq. (4) [16]. That is to say, Eq. (4) is a modified version of the Weibull model. Furthermore, Eq. (4) collapses to the homogeneous model when  $h = 0$  [15].

### 2.2. Classification of the dissolution profiles

As shown in Fig. 1, according to the type of dissolution rate function  $k_t(t)$ , the dissolution profiles can be classified into three non-parametric classes, including declining dissolution rate (DDR), constant dissolution rate (CDR), and increasing dissolution rate (IDR). The differences among these classes are less visible

but can be characterized by the relative dispersion  $CV^2$  [18,20]. For Weibull model, the relative dispersion  $CV^2$  is

$$CV^2 = \frac{2(1-h) \cdot \Gamma\left(\frac{2}{1-h}\right)}{\Gamma^2\left(\frac{1}{1-h}\right)} - 1 \quad (5)$$

where  $\Gamma$  refers to the gamma function [18]. The DDR, CDR, and IDR classes correspond to  $CV^2 > 1$ ,  $CV^2 = 1$ , and  $CV^2 < 1$ , respectively.

### 2.3. The heterogeneity of dissolution process

Dissolution heterogeneity is defined in such a way that the probability to become part of the dissolution process is not identical for all molecules [18]. As a measurement of heterogeneity, the Kullback-Leibler information distance  $d_{K-L}$  is derived using a series of statistical information theory [15,18]:

$$d_{K-L} = \ln \Gamma\left(1 + \frac{1}{1-h}\right) + \ln(1-h) + C \cdot \frac{h}{1-h} \quad (6)$$

where  $C = 0.577215$  is Euler's constant. It's not difficult to understand that  $d_{K-L} = 0$  is equivalent to the homogeneous case. There is no upper limit for  $d_{K-L}$  and it can be any positive number. The higher the value of  $d_{K-L}$  is, the further the dissolution profile deviates from the homogeneous model (first-order kinetic model).

## 3. Materials and methods

### 3.1. Materials

Liquid HLAS (98%), semi-solid NaLAS paste (22% water by weight) and  $\text{Na}_2\text{CO}_3$  powder were supplied by Procter & Gamble Technology (Beijing) Co., Ltd. The NaLAS paste is a highly viscous opaque yellow gel with a viscosity of 800 Pa·s at the shear rate of  $10 \text{ s}^{-1}$  at 298.15 K [4]. It was prepared through the complete neutralization reaction between HLAS and NaOH aqueous solution. HLAS is a brown liquid with a viscosity of 1 Pa·s at the same condition and able to wet the  $\text{Na}_2\text{CO}_3$  particles [21,22]. De-ionized water (DI water) was used in all experiments.

### 3.2. Preparation of detergent agglomerates

The detergent agglomerates were granulated in a lab-scale high-shear mixer (Braun CombiMax, Germany) equipped with a vessel of 2 L, a two-blade impeller and a feed inlet arranged on the top of the unit. Room temperature was adopted to perform the granulation experiments. Preliminary experiments were carried out to select the feasible range of process variables. The results showed that mixing time in the range of 2–3 min and stirring speed of 1000 rpm should be chosen as the optimal conditions, which would be benefit for binder dispersion and particle growth. The batch size was 300 g. In this paper, 14 kinds of formulas were prepared by using two different binders. The chemical compositions are summarized in Table 1.

For type 1, the semi-solid NaLAS paste was employed as binder. The  $\text{Na}_2\text{CO}_3$  powder was added along with NaLAS paste into the mixer, and then the granulation process was started. The addition amounts of  $\text{Na}_2\text{CO}_3$  and binder for each formula are detailed in Table S1 (Supplementary Material). There was no reaction between the NaLAS paste binder and the  $\text{Na}_2\text{CO}_3$  powder during granulation. After mixing for 2–3 min, the agglomerates were collected and dried at 50 °C till the moisture content reached 2%. The moisture content was determined by Halogen moisture analyzer (HR83, Mettler Toledo, Switzerland). Then the agglomerates were sealed in bags and stored in a desiccator. In order to examine the influence of binder content, 7 kinds of formulas for type 1 were prepared with the NaLAS content varied from 20% to 50%.

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