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Prediction of aggregation behavior of submicron-sized particles of praseodymium-doped zirconium silicate in aqueous suspension by population balance model

Libin Liang [a], Yanmin Wang [a,*], Zhidong Pan [a,b]

[a] School of Materials Science and Engineering, South China University of Technology, Guangzhou 510640, China
[b] Guangdong KITO Ceramics Co., Ltd., Foshan 528031, China

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ABSTRACT

The aggregation behavior of submicron-sized particles of praseodymium-doped zirconium silicate, a ceramic pigment, in aqueous suspension was predicted by a modified population balance model. In the model, the collision frequencies were selected to describe evolution of the particle size distribution of the suspension. The collision efficiency was estimated as a function of interaction potential between particles based on Derjaguin–Landau–Verwey–Overbeek theory. The population balance model was modified to predict the stable state of the aggregation by introducing the volume mean size of aggregate to stability ratio. In addition, aggregation of the particles in aqueous suspension in the presence of sodium dodecyl benzene sulfonate or potassium chloride was experimentally investigated. The predicted data (i.e., the final aggregate size, aggregation rate, and particle size distribution) were similar to the experimental results.

Introduction

Ceramic pigments are an integral part of decorative and protective coatings, and are widely used in the ceramics industry as colorants (Cain & Morrell, 2001; Cavalcante, Dondi, Guarini, Raimondo, & Baldi, 2009; De la Luz, Prades, Beltrán, & Cordoncillo, 2013; Luňáková, Trojan, Luxová, & Trojan, 2013). Praseodymium-doped zirconium silicate (Pr–$ZrSiO_4$) is widely used as a yellow ceramic pigment because of its excellent mechanical performance, excellent thermal properties (i.e., high melting point, low heat conductivity, and low expansion coefficient), high chemical stability, and electrical and optical properties (Garrido & Aglietti, 2001; Jang & Kim, 2000; Lemberger et al., 2004; Sancho, Arnal, Pineda, Catala, & Lora, 2005). The problem with using submicron-sized pigments is their aggregation in organic solvent and water because of their high surface area and surface energy (Cavalcante et al., 2009; Nejad, Baghshahi, & Bakhtiari, 2011). Water-based ceramic ink-jet pigments have attracted much attention because of their environmentally friendly features (Akdemir, Ozel, & Suvaci, 2011). Therefore, one of the most important and promising aspects of submicron-sized pigments is control of the stability and final particle size distribution (PSD) of the aqueous suspension, which affects the subsequent color properties and pigment performance, i.e., tribological and mechanical properties, optical properties, and nozzle clogging caused by dispersion stability (Cavalcante et al., 2009; Gardini et al., 2008; Mott & Evans, 1999; Mott, Song, & Evans, 1999; Roda, Guardigli, Russo, Pasini, & Baraldini, 2000).

The population balance model (PBM) is commonly used to describe particle behavior and predict the evolution of the aggregate size distribution with time. Smoluchowski (1917) predicted flocculation to be a rate process with population balance equations, which have subsequently been extensively used to simulate the kinetics of coagulation and flocculation. The model assumes that the aggregation process occurs in two steps: particle transport and subsequent attachment, which are described by the collision frequency factor and the collision efficiency factor, respectively. In some previous studies (Ding, Hounslow, & Biggs, 2006; Lattuada, Wu, Sandkühler, Sefcik, & Morbidelli, 2004; Spicer & Pratsinis, 1996), the kernels were treated as a fitting parameter or a constant to simplify the calculation. However, this would restrict the applicability of the PBM to specific processes (Runkana, Somasundaran, & Kapur, 2006). Numerous researchers have studied the kernels regarding the suspension and particle properties, as reviewed by Liao and Lucas (2010) and Meyer and Deglon (2011). Runkana,

* Corresponding author. Tel.: +86 20 8711 4883.
E-mail address: wangym@scut.edu.cn (Y. Wang).

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Nomenclature

N_i	number density of particles in section i
x_i	pivot particle size of section i, m^3
g	gravitational acceleration, m/s^2
v_i	volume of the particles in section i, m^3
$h_{i,j}$	aggregation frequency, m^3/s
h^*	parameter of aggregation frequency
k_B	Boltzmann constant, J/K
T	absolute temperature, K
R	center-to-center distance between particles, m
V_T	total energy of interaction, J
r_i	radius of particles in section i, m
$\overline{V}$	mean volume of suspension, m^3
A	Hamaker constant of particles across the solvent, J
e	elementary charge, C
z_c	valence of counter ion
c_i	salt concentration
z_i	valence of electrolyte ions
N_A	Avogadro's number
Pe	Peclet number
b_R	parameter with value 5.32
f_R	retardation correction function
a_m	effective monomer size
D_{Sc}	scaling length
$\bar{r}$	volume mean size of aggregates, m
$h_{i,j}^{Br}$	collision frequency due to Brownian motion, m^3/s
$h_{i,j}^{DS}$	collision frequency due to differential sedimentation, m^3/s
V_A	van der Waals energy between particles, J
V_R	electrical double layer repulsion energy between particles, J
V_S	steric effect energy between particles, J
h_0	particle surface–particle surface separation distance, m
W	stability ratio
c	parameter in Eq. (18)
d	particle diameter, m
R_{RRB}	percentage of volume of particles with diameter > d
d_e	size modulus(diameter when R = 36.8%), m
n	uniformity coefficient
$\beta_{i,j}$	aggregation kernel
ω	assigned factor in Eq. (2)
δ	Dirac function
δ_t	adsorbed polymer layer thickness, m
μ	viscosity of the suspension, kg/(m s)
$\lambda_{i,j}$	collision efficiency
ρ_p	density of the particle, kg/m^3
ρ_l	density of the liquid, kg/m^3
ε_0	dielectric constant of vacuum, C/(mV)
ε_r	dielectric constant of solvent, C/(mV)
κ	Debye–Hückel parameter, m^{-1}
ζ	zeta potential on particle surface, V
φ	surface potential on particle surface, V
λ_R	characteristic wavelength of interaction (typically 100 nm)
α_{Sc}	numerical constant
Γ	total amount of polymer adsorbed on a single surface
Γ_0	adsorbed amount at saturation
ϕ_{S_o}	polymer concentration at a single saturated surface
γ	degree of homogeneity

Somasundaran, and Kapur (2004) developed a mathematical model combining the aggregation process with the chemical properties of the suspension by applying Derjaguin–Landau–Verwey–Overbeek (DLVO) theory. Such a modification could take into account the effect of important process variables, such as electrolyte concentration, on aggregation, which was not possible in some previous PBMs. Moreover, most modeling studies (Runkana, Somasundaran, & Kapur, 2005; Runkana et al., 2006; Sandkühler, Lattuada, Wu, Sefcik, & Morbidelli, 2005) focused on the aggregation process leading to gel phases in a short time, thus the finite-size aggregates, while the long-time stability of the suspensions were not discussed. Recently, Atmuri, Henson, and Bhatia (2013) proposed a model considering aggregates as "particles", and consequently allowing the stability ratio to vary as aggregation proceeds. This seems to be the first PBM that is able to predict the steady state of a system. Furthermore, most previous studies characterized the aggregation process by the mean size of the suspension (Lattuada, Sandkühler, Wu, Sefcik, & Morbidelli, 2003; Lattuada et al., 2004; Runkana et al., 2004; Somasundaran & Runkana, 2005) or only predicted the PSD at a stable state (Ahmad, Chong, & Bhatia, 2008; Atmuri et al., 2013; Runkana et al., 2006). Because aggregation is a particle process, the PSD is the most explicit measure of aggregation (Runkana et al., 2004). Hence, an important component of the development of theoretical models for aggregation of submicron-sized suspensions is simulation of PSD evolution with time.

The aim of this study is to propose a suitable approach for PBM to predict the aggregation behavior of submicron-sized Pr–$ZrSiO_4$ particles in aqueous suspensions. This involves selecting the kernel expressions to describe the evolution of the PSD and combining the kernels with the interaction energy between particles and the volume mean size of the aggregates to simulate the steady state of aggregation.

Model development

The PBM is the most informative model for simulating aggregation phenomena in colloidal suspensions. The discretized form of the PBM for aggregation of particles is given by (Smoluchowski, 1917):

$$\frac{dN_i}{dt} = -N_i\sum_{j=1}^{\infty}\beta_{i,j}N_j + \frac{1}{2}\sum_{j=1}^{i-1}\beta_{i-j,j}N_{i-j}N_j, \tag{1}$$

where N_i is the number density of particles in section i and $\beta_{i,j}$ is the aggregation kernel. In Eq. (1), the first term on the right-hand side describes the loss of aggregates because of aggregation with other particles belonging to all size classes and leaving section i. The second term represents the growth because of aggregation of particles belonging to a smaller section.

To solve the PBM, it is necessary to use a numerical solution after discretizing the equation with respect to size into a set of nonlinear ordinary differential equation. Based on the pivot technique (Kumar & Ramkrishna, 1996), the rate of change of particles during simultaneous aggregation is given by the following discretized PBM:

$$\frac{dN_i}{dt} = \sum_{j,\,k}^{j\geq k}\left(1-\frac{1}{2}\delta_{j,k}\right)\omega\beta_{j,k}N_jN_k - N_i\sum_{k=1}^{M}\beta_{i,k}N_k,$$

$$x_{i-1} \leq \left(x_j + x_k\right) \leq x_{i+1} \tag{2}$$

$$\omega = \begin{cases} \dfrac{x_{i+1}-v}{x_{i+1}-x_i}, & x_i \leq (x_j+x_k) \leq x_{i+1}, \\ \dfrac{v-x_{i-1}}{x_i-x_{i-1}}, & x_{i-1} \leq (x_j+x_k) \leq x_i, \end{cases}$$

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