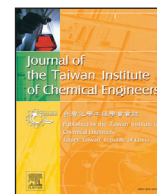




Contents lists available at ScienceDirect

Journal of the Taiwan Institute of Chemical Engineers

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

Theoretical and experimental investigation of time-varying properties in the coagulation of kaolinite containing wastewater by gypsum

Huaigang Cheng^a, Yunyun Zhang^a, Xin Wang^a, Fangqin Cheng^{a,*}, Jing Han^a, Jing Zhao^b, Nan Wang^a, Yang Sun^a

^aInstitute of Resources and Environmental Engineering, State Environment Protection Key Laboratory of Efficient Utilization of Coal Waste Resources, Shanxi University, Taiyuan 030006, China

^bCollege of Chemical Engineering, Qinghai University, Xining 810016, China

ARTICLE INFO

Article history:

Received 28 March 2016

Revised 1 November 2016

Accepted 17 December 2016

Available online xxx

Keywords:

Gypsum

Kaolinite

DLVO model

Ostwald ripening

Coagulation

Time-varying property

ABSTRACT

Besides the well-known mechanism of compressed double electric layer, this paper quantitatively found that the time-varying properties of gypsum particles play an important role on coagulation of kaolinite loaded wastewater. The Ostwald ripening equations were embedded into the DLVO model, based on which we investigated how the size and ξ potential of gypsum particles and the interaction energy between gypsum and kaolinite particles work as a function of time. Four stages were presented when gypsum particles were added into water: the trends in size were smaller, larger, constant and smaller whereas the ξ potential absolute values increased, decreased and remained constant, respectively. The electrostatic and the total potentials between gypsum and kaolinite particles decreased first, followed by an increase–decrease fluctuation as time passed. According to the potential changing, the biggest coagulation tendency happens when the gypsum particles have been put into water for 6–7 min, but it is not stable until the water soaking time reaches 30 min. This deduction was confirmed by the pilot sedimentation test of wastewater with gypsum as a coagulant. Furthermore, the network structures formed by fine ripening gypsum particles were observed and the flocculation effect was confirmed to be another part of the co-sedimentation mechanisms.

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1. Introduction

Clay particle (especially the fine particle, commonly the kaolinite and etc.) suspended in wastewater is one of the sources of environmental pollution. The suspended particles can reduce the transparency of water and the toxic substances adsorbed on the particles may directly kill aquatic organisms. One of the effective ways to treat such wastewaters is the coagulation using gypsum (calcium sulfate dihydrate, $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) [1,2] powder which is harmless to the soil and does not produce secondary pollution and is easy to get in large quantities, although eutrophication may still occur. The compression of electric double layer of clay particles by the gypsum released Ca^{2+} in water is one of the main causes of coagulation. Besides that, the solid gypsum particles can greatly enhance the coagulation process [3], but the detailed information on how it works has not been fully understood. In particular, the gypsum has been observed always changing its size

and number density in water [3], which makes its physicochemical property and coagulation ability time-varying. Perhaps because this phenomenon is strange, to the best of our knowledge, little literature has discussed its mechanism.

Many time dependent phenomenon [4] exist in the coagulation process. From the perspective of practical application, it is necessary to understand the time-varying nature when the gypsum is used in the coagulation technology. The morphology of gypsum is frequently changing in the water. For example, the needle gypsum whisker is formed due to its self-confinement and hydration [5] features. Then, the clay particles or aqueous Ca^{2+} are likely to adsorb on the whiskers, leading to the desliming or desupersaturation of Ca^{2+} in water and the scale prevention [6] (e.g., in precipitator [7] or on membrane [8]). As a result of all these factors, even the concentrated saltwater [9–11] can be deeply treated. At the micro level, the polynucleation [12] and the adsorption [13,14] of ions or CaSO_4^0 ion pairs on particle surface control the gypsum precipitation. This method can be called chemically-enhanced seeded precipitation [14] or accelerated precipitation softening demineralization [8], in which the seed size and the growth kinetics of gypsum are found to be influencing factors [15,16]. Similarly, it has been

* Corresponding author.

E-mail addresses: chenghg@sxu.edu.cn (H. Cheng), cfangqin@sxu.edu.cn (F. Cheng).

<http://dx.doi.org/10.1016/j.jtice.2016.12.021>

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List of symbols

$\langle r_0 \rangle$	average radius of particles at the starting time
$\langle r_t \rangle$	average radius of particles at time t
A_{132}	Hamaker constant interacting between particle 1 and 2 in medium 3
c	concentration
c_∞	total solubility of calcium components
c_s	solubility
D	diffusion coefficient
H	particle center distance between two particles
H_0	surface distance between two particles
h_0	decay length
k	chemical equilibrium constant
k_D	dissolution rate constant
m	molar concentration
NTU	Nephelometric Turbidity Unit
p	parameter for calculation in the DLVO model
q	parameter for calculation in the DLVO model
q_t	amount of surface charge
R	particle radius
R_g	the ideal gas constant
r_t	radius of particle
S_t	specific surface area
T	absolute temperature
t	time
V	volume
V_E	electrostatic energy
V_H	polarization energy
V_H^0	energy constant of interface polar interactions
V_T	total potential energy
V_W	Van der Waals interaction energy
Greek letters	
μ	electrophoretic mobility
a	activity
γ	activity coefficient calculated using Debye–Hückel equations
γ_s	surface tension
ε_a	absolute dielectric constant of dispersed medium
η	viscosity
κ^{-1}	reciprocal of Debye length
ξ	ξ potential
ν	molar volume of calcium components in the solution
Φ	the volume fraction of particles in suspension
ψ	surface potential of a particle

found that the time-varying properties for preparation of coagulant (aging time) has great effect on the coagulation performance [17].

From the aspect of theoretical research, the DLVO (Derjaguin–Landau–Verwey–Overbeek) model [18–20] is a classic theory that describes the systems of suspended particles. However, the dynamic process cannot be directly described using the DLVO model due to the absence of the time term in its equations. In order to solve this problem, the combination of the DLVO model with other research methods may be tried. In fact, this thought has been used to expand the application of DLVO model. For example, statistical methods or mathematical models are used to calculate the surface features of particles that are embedded into the DLVO model. For the dispersion of bitumen droplets in water, the surface charge and interaction forces between droplets could be calculated when the ξ potential is regarded as a random variable in accordance with the Gaussian distribution. Then, the probabilities of coalescence

between droplets could be predicted using the DLVO model [21]. This similar method was used to investigate the interaction force between particles and a wall [22], thus calculating the forces of attraction and repulsion at different locations based on the random distribution of nanoscale chemical and topographical heterogeneities. Similar method may be considered to solve the problem of potential variation with time, because the DLVO model cannot determine the potential change with time but the use of other models is possible.

Generally, the crystals in different growth stages have different coagulation effects, so the changing behavior of the surface properties of gypsum is important to water purification. Based on the above analysis, we propose describing the dispersion of gypsum and fine clay particle system by calculating the real-time surface characteristics using other models and embedding them into the DLVO model, which is the objective of this work.

2. Material and methods

Analytically pure grade gypsum (Tianjin Bodi Co. Limited, China) was used and the kaolinite (Yunnan Tianhong Co. Limited, China) were examined by XRD (Bruker D2 Phaser, Germany) to be 95% in purity. Deionized water was prepared using a Milli-Q water purifier (Millipore Corp 18 M \sim cm). The gypsum was ground to an average size of 96–109 μ m and added to deionized water or kaolinite-loaded deionized water. Then the average particle size, particle size distribution, ξ potential and turbidity in the suspensions were measured in real time. A pH meter (PHS-3C, Shanghai REX Instrument Factory) was used to measure pH values. Turbidity was measured using a turbidimeter (HACH 2100AN, USA). The particle size distribution was determined using a particle monitor (Mettler FBRM G400, Switzerland) and a particle size analyzer (ANKERSMID EyeTech, Holland). ξ potential was determined using a zetasisizer (Malvern NanoZS90, UK). An optical microscope (Nikon ECLIPSE 50i, Japan) were used to observe particle morphology.

Furthermore, a pilot test was conducted, as shown in Fig. 1. Gypsum was screened and transferred to tanks ($\Phi 2000 \times 2000$ mm in diameter \times height) and ripened for different times. Then, the wet gypsum was mixed into kaolinite suspensions in a sedimentation pond. Intermittent experiments were conducted. Both the gypsum-loaded water and the kaolinite-loaded water were 3 m³ in volume in each of the experiments. The depth of mixed suspensions was approximately 200 mm in the sedimentation pond and turbidity was tested 100 mm beneath the water surface.

3. Extended DLVO theory with changing particle size

The total potential energy, V_T , controls coagulation or dispersion between particles according to the extended DLVO theory, including the Van der Waals interaction energy, V_W the electrostatic energy V_E and the polarization energy V_H between particles [23]. The particle's repulsion and dispersion is shown by a positive V_T value, and *vice versa*. As to the spherical particles with radii R_1 and R_2 ,

$$V_W = -\frac{A_{132}}{6H} \cdot \frac{R_1 R_2}{(R_1 + R_2)} \quad (1)$$

$$V_E = \frac{\pi \varepsilon_a R_1 R_2}{R_1 + R_2} (\psi_{01}^2 + \psi_{02}^2) \left[\frac{2\psi_{01}^2 \psi_{02}^2}{\psi_{01}^2 + \psi_{02}^2} p + q \right] \quad (2)$$

$$V_H = \frac{2\pi R_1 R_2}{R_1 + R_2} h_0 V_H^0 \exp\left(\frac{H_0 - H}{h_0}\right) \quad (3)$$

where H and H_0 are the particle spacing and the surface distance of the two particles, respectively. A_{132} is the Hamaker constant interacting between particles 1 and 2 in medium 3. $p = \ln\left[\frac{1+\exp(-\kappa H)}{1-\exp(-\kappa H)}\right]$ and $q = \ln[1 - \exp(-2\kappa H)]$. ψ_{01} is the surface

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