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Short communication

Stability evaluation of aqueous alumina–zircon–silicon carbide suspensions by application of DLVO theory

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Abstract

The colloidal stability of alumina–zircon–silicon carbide suspensions has been investigated by measuring particle size distribution, sedimentation height and zeta potential. A polyelectrolyte (Dolapix CE-64) was used as a dispersant in different concentrations. Silicon carbide (SiC) particles lowered the zeta potential of suspensions, sedimentation height and also average particle size of resultant powders. It was concluded that the stability of alumina–zircon suspensions reaches an optimum condition by adding 20 vol.% SiC particles when using 0.5 wt.% dispersant, however, further increasing of SiC particles reduces the stability of suspensions. In this overview we present the basic theory for the double layer near the surface of alumina particles. Good agreement was found between dispersion characteristics of suspensions from theory and experimental results.

Keywords: B. Interfaces; Composite materials; Adsorption; Surface properties

1. Introduction

In colloidal processes, it is important to evaluate the dispersion state of powders in liquid [1]. Density and microstructure of green and sintered compacts can be better controlled in colloidal process rather than conventional dry pressing [2–4]. When particles have the same surface charges in suspension, the electrostatic repulsion between particles may cause them to be dispersed. It is important to know the behavior of the particles in suspensions [5,6]. In the previous work [7] the effect of additions of SiC particulates on rheological and sintering behavior of slip-cast alumina–zircon (AZ) composites was investigated. Results showed that the use of polyacrylate dispersant up to 0.5 wt.% creates the stable aqueous suspensions containing 40 vol.% solid.

In the present work, the effect of SiC content and various amounts of Dolapix on the average size of particles and resultant potential energies of alumina–zircon suspensions was examined and tried to analyze the suspension behavior by DLVO theory.

2. Experimental procedures

The powders used in this work were α-alumina (MR70, Martinswerk, Germany, 0.6 μm), zircon (Zircosil, Johnson-Matthey, Italy, 1.4 μm) and SiC particles (Mirali, Iran, 4 μm). Dolapix CE-64 (Zschimmer & Schwarz) was used as a dispersant. Aqueous alumina–zircon suspensions with 40 vol.% solid were prepared by adding the required weight fractions of alumina and zircon powders (85/15 wt.%.) in distilled water. Dolapix was used at various amounts (0.3, 0.5 and 0.7 wt.% based on the solid weight). Slips were prepared by mixing the dispersant with water and subsequent pouring of mixed powders in the dissolved dispersant. Slurries were stirred for 20 min by a planetary mill [7]. Zeta potential and particle size of suspensions were determined by Zetasizer3000 HS_A (Malvern) and by particle size analyser (Fritsch analysette, Germany), respectively. The pH of all suspensions was kept at 9–9.5. Since the most part of particles in prepared suspensions were alumina and the main adsorption of Dolapix occurred on the surface of these particles, then for simplicity the theoretical

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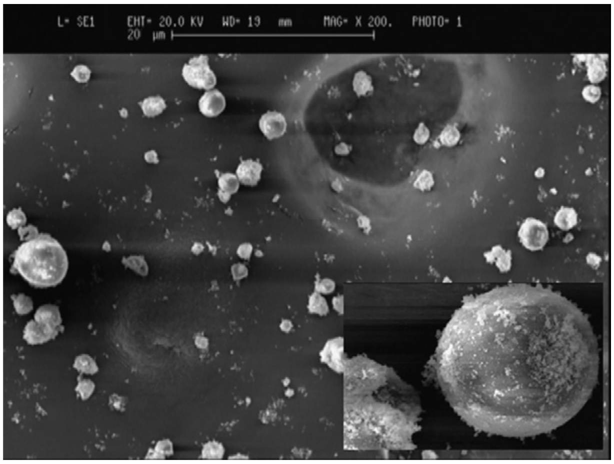

Fig. 1. SEM micrograph of alumina powder.

calculations were first carried out on the spherical like alumina particles. Fig. 1 shows a typical SEM (S360 Cambridge) micrograph of spherical alumina particles. The stability measurements of suspensions were carried out using the conventional sedimentation technique in a graduated cylinder [8]. Lower sedimentation height indicates that the settling rate is slow and suspension is stable.

3. Results and discussion

3.1. Zeta potential measurements

Table 1 shows the zeta and surface potential of alumina–zircon–SiC (AZS) suspensions. Table 2 also shows the zeta potential of suspensions that have been measured as a function of Dolapix percent. There is a considerable increase in zeta potential of AZ20S suspension by adding 0.5 wt.% dispersant. The minimum zeta potential values presented in Table 2 determine the optimum dosage of Dolapix in each suspension. Since Dolapix is completely dissociated above pH 8.5 [9], therefore, it assumes that only Dolapix monovalent ions (COO^- and NH_4^+) are present in solution. With mentioned assumption, $\kappa = 0.64\ nm^{-1}$ ($\kappa^{-1} = 1.56$ nm) was calculated from appropriate equation [10]. Potential curves of AZS suspensions were calculated as a function of Dolapix content (Fig. 2) [11]. The observed behavior is as the result of adsorption or desorption of ionic species in solution (such as dissociated Dolapix). Fig. 2a shows that the increase of SiC particles in AZ suspension leads to increase the surface charge of particles (as proved by increasing the negative values of zeta potential). The higher surface charge serves the higher repulsive forces between the

Table 1
Zeta potential and surface potential of AZS suspensions.

SiC (vol.%)	Zeta potential (mV)	Surface potential (mV)	Reference code
0	−25 ± 1	−68	AZ0S
10	−31 ± 1	−84	AZ10S
20	−48 ± 2	−130	AZ20S
30	−33 ± 1	−90	AZ30S

Table 2
Zeta potential of AZS suspensions as function of Dolapix (±1).

Dolapix (wt.%)	AZ0S	AZ10S	AZ20S	AZ30S
0.3	−24	−29	−30	−26
0.5	−25	−31	−48	−33
0.7	−23	−24	−27	−29

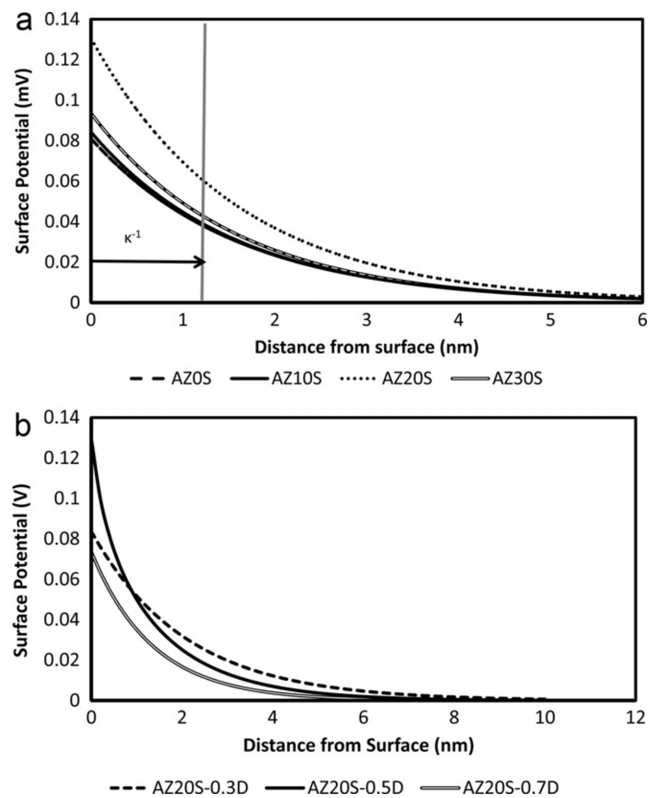

Fig. 2. Potential curves of (a) AZS suspensions and (b) AZ20S suspension as a function of Dolapix percent.

particles and subsequently a stable suspension can be obtained. The zeta potential measurements showed that the potential and stability of suspensions containing 20 vol.% SiC are high. Fig. 2a further reveals that AZ30S suspension is less stable than AZ20S suspension due to a lower surface potential in the former suspension. Adsorption of negatively charged carboxylic group dissociated from Dolapix on the surface of alumina particles via electrostatic attraction results in more alumina charged particles [1]. It can be seen that by adding 0.5 wt.% Dolapix, the surface of particles will be more negatively charged which subsequently brought more stability (Fig. 2b). Increasing the concentration of Dolapix up to 0.7 wt.% resulted in increase of the steric hindrance and electrostatic repulsion between Dolapix chains. These two repulsive forces mentioned above further lead to the incomplete surface coverage and decreased in the net negative charge of the powders due to the excessive amounts of dispersant [12].

3.2. Sedimentation test

The result of sedimentation measurements of suspensions containing different fractions of SiC particles is shown in Fig. 3.

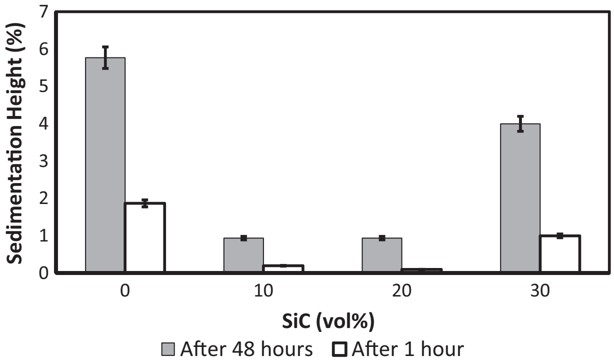

Fig. 3. Sedimentation height of alumina–zircon suspensions versus SiC concentration.

As usually known, as the sedimentation height is lowered in a suspension, it will be more stable [4]. As can be seen, by increasing the SiC content, the sedimentation height percent decreases significantly and reaches a minimum of 0.94% at 20 vol.% SiC (AZ20S). By increasing the SiC content over 20 vol.%, the sedimentation height increased dramatically which can be explained by the fact that the zeta potential and subsequently electrostatic repulsion between particles decrease by increasing the SiC content, as can be observed for AZ30S suspension.

3.3. Particle size distribution

Fig. 4 shows the average particles sizes of suspensions as a function of SiC content. From 0 to 10 vol.% SiC, the average particle size in suspension decreases gradually and then increases rapidly up to 1.84 μm by increasing SiC content to 30 vol.%. The increase of particle size in AZ30S suspension can be confirmed by the reduction of zeta potential values.

3.4. Calculated interaction energy

Fig. 5 shows the calculated [13] total energy of alumina particles in AZS suspensions. The calculated DLVO interparticle potentials between alumina particles have shown that the repulsive forces are dominant interparticle forces in AZ20S and AZ30S suspensions (Fig. 5a). This leads the formation of deflocculated suspensions with a high stability. Although AZ30S suspension has higher interparticle repulsion energy than AZ10S suspension, it also has a higher *d*50 value. High *d*50 value means rapid settling and fast sedimentation of particles in suspension. Therefore, it can be said that the high repulsive energy of particles in AZ30S is not sufficient to hinder the sedimentation of particles and lead to an unstable suspension. Furthermore, the DLVO interparticle interaction potentials of AZ20S suspension as a function of Dolapix content were calculated and the results are shown in Fig. 5b. It can be observed that the highest energy barrier belongs to AZ20S suspension containing 0.5 wt.% Dolapix. Therefore, the optimum dosage of dispersant Dolapix to achieve maximum dispersion of AZS suspensions was determined as 0.5 wt.%.

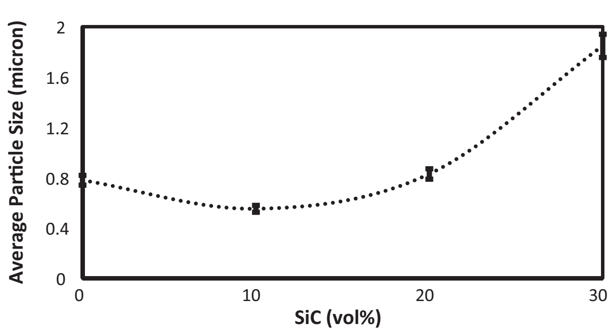

Fig. 4. Average particle size of AZS suspensions.

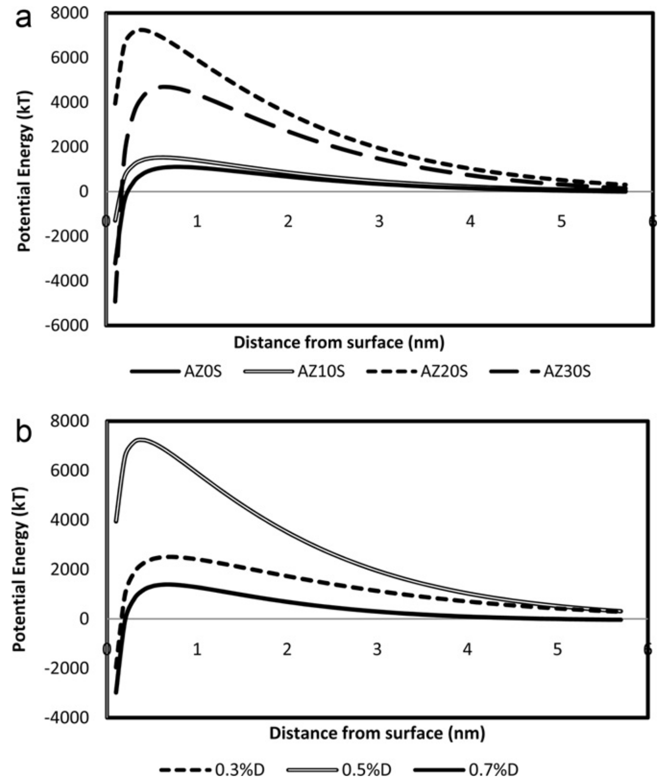

Fig. 5. Total energy of (a) AZS suspensions, (b) AZ20S suspension as a function of Dolapix percent.

In order to quantify the effect of SiC on the alumina–zircon system and realize the DLVO theory for the entire system, each particle interaction needs to be specifically analyzed: alumina–alumina, zircon–zircon, zircon–alumina, SiC–alumina, etc. Repulsion energies between alumina–zircon, alumina–SiC, zircon–zircon, zircon–SiC and SiC–SiC have also been calculated (Fig. 6). It can be observed that the maximum value of repulsion energy is between SiC–SiC particles. Using SiC particles can increase the repulsion energy between other particles too.

3.5. Stability ratio

Stability of any dispersion can be easily determined by one of the useful quantitative methods named as the stability ratio (*W*) [14]. The higher value of W means the higher stability of a suspension [15]. Energy barrier and log *W* of AZS suspensions

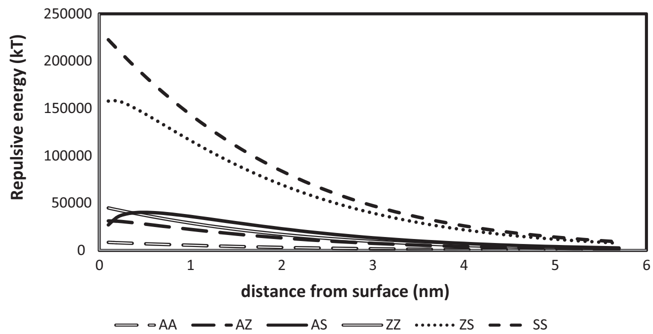

Fig. 6. Repulsion energies between alumina–alumina (AA), alumina–zircon (AZ), alumina–SiC (AS), zircon–zircon (ZZ), zircon–SiC (ZS) and SiC–SiC (SS) components.

Table 3
Energy barrier and log *W* of AZS suspensions.

SiC (vol.%)	0	10	20	30
G_{max} ($\times 10^{-17}$ V)	0.45	0.63	2.98	1.93
log *W*	0.48	0.96	3.01	0.89

Table 4
Energy barrier and log *W* of AZ20S suspensions as function of Dolapix.

AZ20S-Dolapix (wt.%)	0.3	0.5	0.7
G_{max} ($\times 10^{-17}$ V)	1.03	2.98	0.57
log *W*	1.04	3.01	0.58

were calculated and the results are presented in Tables 3 and 4. As a result, AZ20S suspension containing 0.5 wt.% Dolapix has the highest stability.

3.6. Interaction of Al_2O_3 with Dolapix

The dispersion of a suspension using Dolapix CE-64 is carried out through electrosteric stabilization [9]. The authors suggest the schematic adsorption of Dolapix on the surface of alumina particle as presented in Fig. 7. As expected, alumina particles at pH 9–9.5 are slightly positively charged. Therefore, it is expected that the negatively charged carboxylic group dissociated from dispersant is adsorbed on the positively charged alumina surface and consequently the initial surface becomes negatively charged. By adding SiC particles to the suspensions, the negatively charged surface of SiC particles repels the negatively charged surface of alumina particles and the resultant suspension remains stable. AZ suspensions can be stabilized by introducing the SiC particles which increased the net negative charge and zeta potential values. The size of particles increases in suspension which has higher SiC content due to large particles. The increase of SiC particles to 30 vol.% makes AZ suspensions unstable because of increasing the size of particles in suspension which subsequently increases the sedimentation height.

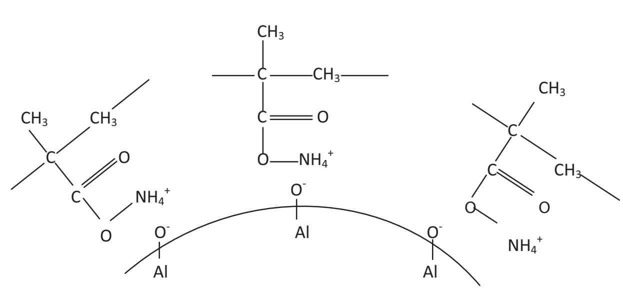

Fig. 7. Schematic adsorption of Dolapix on the alumina surface.

4. Conclusions

This work presents theoretical evaluation of dispersion behavior of alumina–zircon–SiC suspensions in the presence of dispersant Dolapix CE-64. Results showed that the stability of alumina–zircon suspensions increased by adding SiC particles up to 20 vol.% as a result of increasing negative zeta potential and lowering sedimentation height and also particle size distribution. Dolapix CE-64, a commercial carbonic acid based polyelectrolyte, was found to be a suitable dispersant for alumina–zircon–SiC suspensions. The Dolapix amount in suspension greatly influences the dispersibility of particles. The use of 0.5 wt.% Dolapix increased the interparticle interaction potential, surface charge (more negative zeta potential) and stability of suspensions. Furthermore, DLVO theory was correlated well with other experimental properties of prepared suspensions.

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