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# Effect of SiC particles on rehological and sintering behaviour of alumina–zircon composites

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**Abstract**

The effect of additions of SiC particulates on rheological and sintering behaviour of slip-cast alumina–zircon composites has been investigated. Finely divided alumina, zircon and silicon carbide powders were first processed into slips, using polyacrylite dispersant (0.5 wt.%) to create highly concentrated, stable aqueous suspensions at 40 vol.% loadings, from which test specimens which were then slip cast and dried. They were subsequently sintered in air for 2 h at 1650 °C. Rheological properties of the prepared slips were evaluated and related to the amount of added SiC. After sintering, the resultant porosities, fractional densities, crystallographic phases present, and microstructures were determined.


*Keywords:* A. Suspensions; A. Solid state reaction; A. Sintering; A. Slip casting; Alumina–mullite–zirconia–SiC composites

## 1. Introduction

Ceramic matrix composites have found increasing applications as structural components where high temperature strength, wear and corrosion resistance are required. Alumina–mullite–zirconia composites (AMZ) are the particular class among the various composites which are widely used for producing refractory parts in glass and refractory industries [1–3]. The AMZ composites have been prepared in two ways, hot-pressing and slip casting [1]. The preparation of these composites using zircon instead of zirconia has attracted the attention of the scientists since the zircon powder with a good quality is ready available from beach sand with a cheap price, thus making the development cost effective [4]. Although conventional dry preparation methods have been used to produce composites, there are few available works on the casting method of these composites. On the other hand, the colloidal processing has been realized to be useful for forming ceramic green bodies [5]. The powder mixture must be dispersed in water to achieve optimum particle packing through colloidal processing. This requires the preparation and dispersion of highly concentrated suspensions which are fundamental steps in the colloidal processing of ceramics [6]. In order to have a successful colloidal processing, polyacrylate based dispersants have been commonly used to improve and control the colloidal stability and rheological properties of concentrated ceramic suspensions. Colloidal stability of concentrated suspensions using polyelectrolytes was attributed to both electrostatic repulsion and steric stabilization mechanism. The adsorption of anionic polyelectrolytes on the powder surfaces increases the negative surface charge and produces some steric repulsion [4].

In the present work, the rheological behaviour of highly concentrated suspensions containing alumina, zircon and SiC particles in the presence of a polyelectrolyte was studied. The rheological measurements were based on the effect of dispersant content, solid loading, pH values and volume fraction of SiC particles on viscosity, shear rate and shear stress.

## 2. Experimental procedures

### 2.1. Materials

The α-alumina (MR70, Martinswerk, Germany), zircon (Zircosil, Johnson-Matthey, Italy) and SiC (Mirali Company, Iran) powders with mean particle size of 0.6, 1.4 and 4 μm, respectively, were used as the starting materials. Dolapix CE-64

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(Zschimmer and Schwarz), which is a polyelectrolyte and acts through electrosteric stabilization mechanism, was used to disperse aqueous alumina–zircon suspensions.

### 2.2. Suspension preparation and rheology

The mass ratio of alumina to zircon in all mixtures was 85/15. Aqueous alumina–zircon suspensions at 40 vol.% solid loading were prepared using different amounts of Dolapix (0.3–0.7 wt.% based on solid weight). Each slip was prepared by solving of the dispersant in water and subsequent pouring of mixed powders in the dissolved dispersant. The resultant slurry was stirred for 1 h and then ultrasonically agitated for 3 min.

Rheological measurements were performed to determine the stability of suspensions using viscometer Physica MCR300 of coaxial cylinders at 25 °C. Zeta potential of suspensions was determined by Zetasizer3000 $HS_A$ (Malvern). The adjustment of pH was achieved with NaOH or HCl. The amount of dispersant adsorbed on particles surfaces was measured using titration method [7].

### 2.3. Sample preparation and sintering

The composite powders containing 0, 10, 20 and 30 vol.% SiC were stabilized and the resultant suspensions were consolidated through slip casting method using plaster moulds. The cast parts were healed at room temperature for 24 h, dried at 110 °C in an oven and finally sintered at 1650 °C for 2 h.

### 2.4. Physical properties and characterizations

Density and porosity of sintered samples were determined by the water adsorption method. Crystalline phases in fired samples were characterized by XRD (Siemens, D500 system) using Cu Kα radiation working with 30 kV accelerating voltage. The microstructure of sintered samples was observed by SEM (VEGA II, TESCAN company) on polished surfaces which were thermally etched at 1500 °C for 20 min.

## 3. Results and discussion

### 3.1. Effect of dispersant concentration on the rheological behaviour

Fig. 1 shows the variation of apparent viscosity as a function of dispersant content. The well-dispersed suspensions containing 0, 10, 20 and 30 vol.% SiC were prepared using 0.5 wt.% dispersant. As Fig. 1 further reveals, in the absence of SiC particles the minimum viscosity (40 mpa s) appears at 0.5 wt.% dispersant. It is observed that the viscosity of suspensions increases with decreasing the dispersant content which can be attributed to an incomplete covering of particles surfaces with polyelectrolyte. Additionally, with further decreasing in dispersant content, the net negative surface charge of all particles decreases and this means that the heterocoagulation mechanism (electrostatic attraction between two or more particles with opposite charge) would be more probable. In the absence of SiC particles, the apparent viscosity gradually increased with increasing the dispersant content higher than 0.5 wt.%. This occurs most probably because of immersing the excessive dispersant into the medium. Free electrolytes in a suspension disturb the electrostatic forces within the particles and decrease the amount of available solvent and subsequently increase the viscosity of the suspension [8]. As reported [6], the existence of free polyacrylate has a detrimental effect on the stability of a suspension with the promotion of the flocculation. The above effect extends with an increase in the molecular weight of polymer.

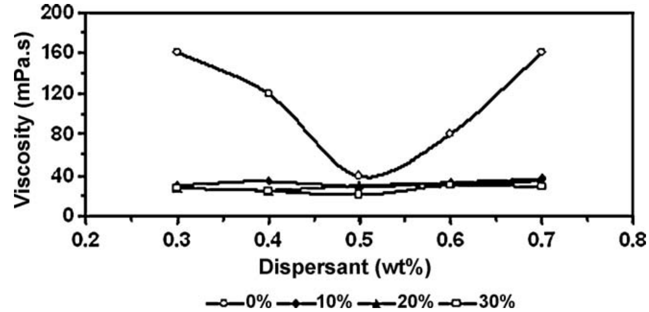


Fig. 1. Viscosity variations of alumina–zircon suspensions with and without SiC particles (vol.%) as a function of Dolapix.

### 3.2. Effect of pH on the rheological behaviour

Fig. 2 demonstrates the apparent viscosity of alumina–zircon suspensions with and without adding SiC particles as a function of pH. It is observed that suspensions dispersed with 0.5 wt.% dispersant indicate a low viscosity at pH 9 and as pH value goes higher the viscosity increases. As reported [9–13], the isoelectric point of alumina, zircon and SiC is at the pH around 8.5, 5.5 and 5, respectively. From these pH values, it can be postulated that the electrostatic interaction between SiC, zircon and alumina particles in suspension is attractive within the pH range 5–9. Based on the explanation mentioned above, alumina–zircon–SiC suspensions should be prepared at a high pH ($pH > 9$), whereas the degree of dissociation of polyacrylate in the pH 9 is higher than that in the pH 7 [14]. The

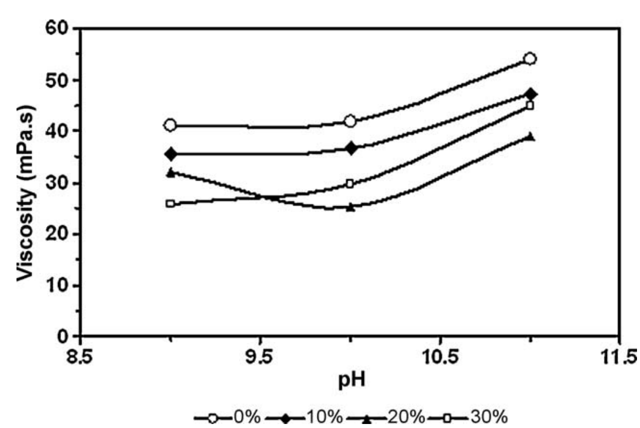


Fig. 2. Viscosity variations of alumina–zircon suspensions with and without SiC (vol.%) as a function of pH.

weak adsorption of dispersant on surfaces at a high pH (e.g. pH 11) may be attributed to the presence of few positive surface sites.

### 3.3. Zeta potential measurements

The zeta potential measurements for different suspensions dispersed with 0.5 wt.% dispersant are given in Table 1. The suspensions prepared from individual alumina, zircon and SiC particles were well-dispersed and showed highly negative zeta potential values using 0.5 wt.% dispersant because of their significant surface charges. As Table 1 further reveals, the alumina–zircon suspension has a higher zeta potential value than each suspension produced from individual powders. It is also observed that alumina suspension becomes more stable by adding zircon particles because of increasing the net negative surface charges. As Table 1 further reveals, as more SiC particles were added into the alumina–zircon suspensions, the higher values of zeta potential obtain. The flow curves of suspensions with 40 vol.% solids loading and different volume fractions of SiC are shown in Fig. 3. A shear thinning behaviour can be observed from Fig. 3a particularly for high volume fraction of SiC particles. This is most probably caused by decreasing the volume fraction of fine alumina particles and subsequently the increase of SiC particles in the initial solids loading. In this case, the increment of zeta potential values occurs with increasing SiC content, since at pH 9, the higher net negative charged surfaces appear for SiC particles than alumina particles. The repulsion forces between the interference particles cause the increase of negative charged surfaces and eventually produce a suspension with a low viscosity. On the other hand, as the volume fraction of large SiC particles increases the sedimentation rate increases and eventually the resultant suspension is diluted and behaves watery.

Table 1
Zeta potential of suspensions at pH 9.

| Suspension | Zeta potential (mV) |
|---|---|
| Alumina + 0.5 wt.% Dolapix | −21.1 |
| Zircon + 0.5 wt.% Dolapix | −38 |
| SiC + 0.5 wt.% Dolapix | −39.97 |
| Alumina–zircon–0% SiC suspension (0.5 wt.% Dolapix) | −44.8 |
| Alumina–zircon–10% SiC suspension (0.5 wt.% Dolapix) | −47.5 |
| Alumina–zircon–20% SiC suspension (0.5 wt.% Dolapix) | −49.72 |
| Alumina–zircon–30% SiC suspension (0.5 wt.% Dolapix) | −46 |

### 3.4. Effect of SiC content on the rheological behaviour

Fig. 3 reveals that the alumina–zircon suspensions exhibit pseudoplastic behaviour with adding SiC particles. This means that the viscosity decreases with increasing the shear rates. The effect of solids loading on rheological behaviour of suspensions containing 20 vol.% SiC was also examined and results are presented in Fig. 4. By increasing the solids loading, the viscosity tends to increase as the result of reducing the particle distances and increasing the frequency of collisions (Fig. 4a). The shear thinning characteristic of alumina–zircon suspensions containing 20 vol.% SiC (Fig. 4) also confirms the complex structure of the resultant suspensions with increasing the solid content [15]. The rheological behaviour of suspensions containing 10 and 30 vol.% SiC (not shown) exhibited a similar behaviour as shown in Fig. 4. As Fig. 5 reveals, the addition of SiC to alumina–zircon suspensions leads a decrease in adsorption of dispersant. This behaviour can be explained by the fact that the positive sites and subsequently adsorption of

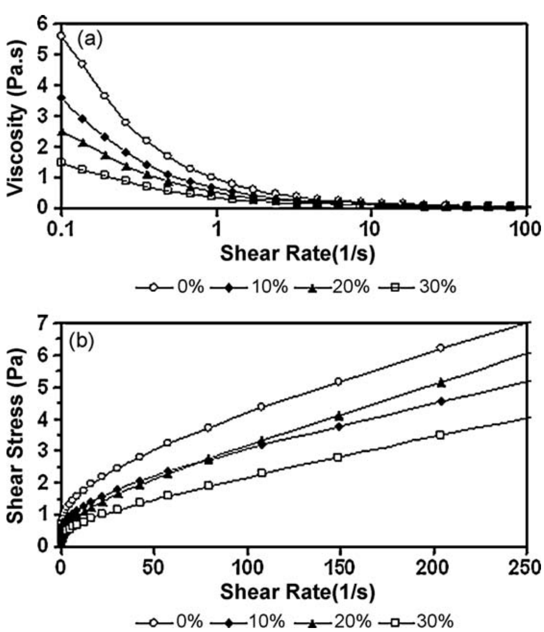


Fig. 3. Flow curves of alumina–zircon suspensions for different amount of SiC (vol.%), (a) viscosity vs shear rate and (b) shear stress vs shear rate.

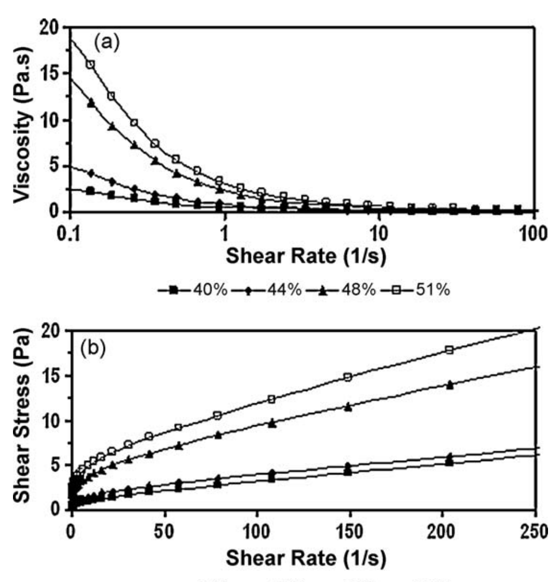


Fig. 4. Flow curves of alumina–zircon–20 vol.% SiC suspensions for different solid loading (a) viscosity vs shear rate and (b) shear stress vs shear rate.

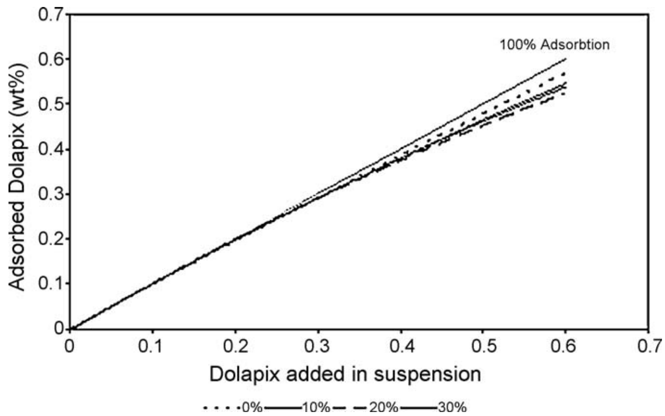


Fig. 5. Adsorption of Dolapix in alumina–zircon–SiC suspensions for different SiC volume percent.

dispersant decrease at pH 9 by increasing the highly negative SiC particles.

### 3.5. Phase and morphological characterizations

The X-ray patterns of sintered samples are shown in Fig. 6. No zircon peaks were detected while zirconia phases were found in both monoclinic and tetragonal types. α-alumina, mullite, monoclinic- and tetragonal-zirconia, and SiC were detected as the only crystalline phases in all sintered composite samples. Fig. 7 shows the relevant density and apparent porosity of composite samples sintered at 1650 °C after 2 h with changing SiC content. As can be seen, the addition of SiC reduces density and increases porosity of the samples. From 0 to 30 vol.% SiC, density decreases 15.8% while porosity increases 96.7%. The SEM image of sintered samples reveals a microstructure with compacted particles in sample without SiC (Fig. 8a) while a porous microstructure is observed by adding 20 vol.% SiC (Fig. 8b). The addition of SiC particles results in an increase in the average grain size of the composite samples so that the average grain size of the sample with 20 vol.% SiC was 1.9 μm while this value decreases to 1.3 μm for sample with no SiC.

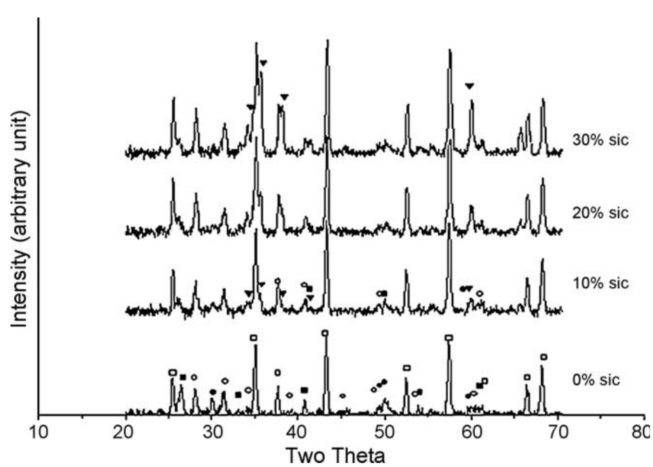


Fig. 6. XRD patterns of samples sintered at 1650 °C for 2 h (■: mullite, ●: tetragonal-zirconia, ○: monoclinic-zirconia, □: alumina, ▼: SiC).

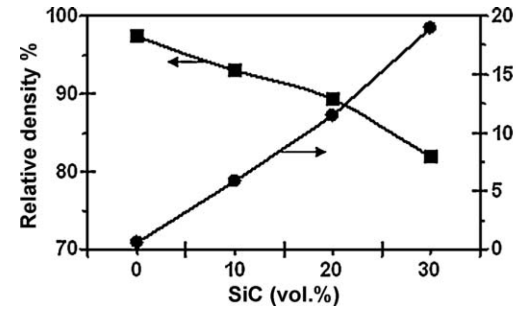


Fig. 7. The change of relative density and apparent porosity of samples sintered at 1650 °C for 2 h by adding SiC particles.

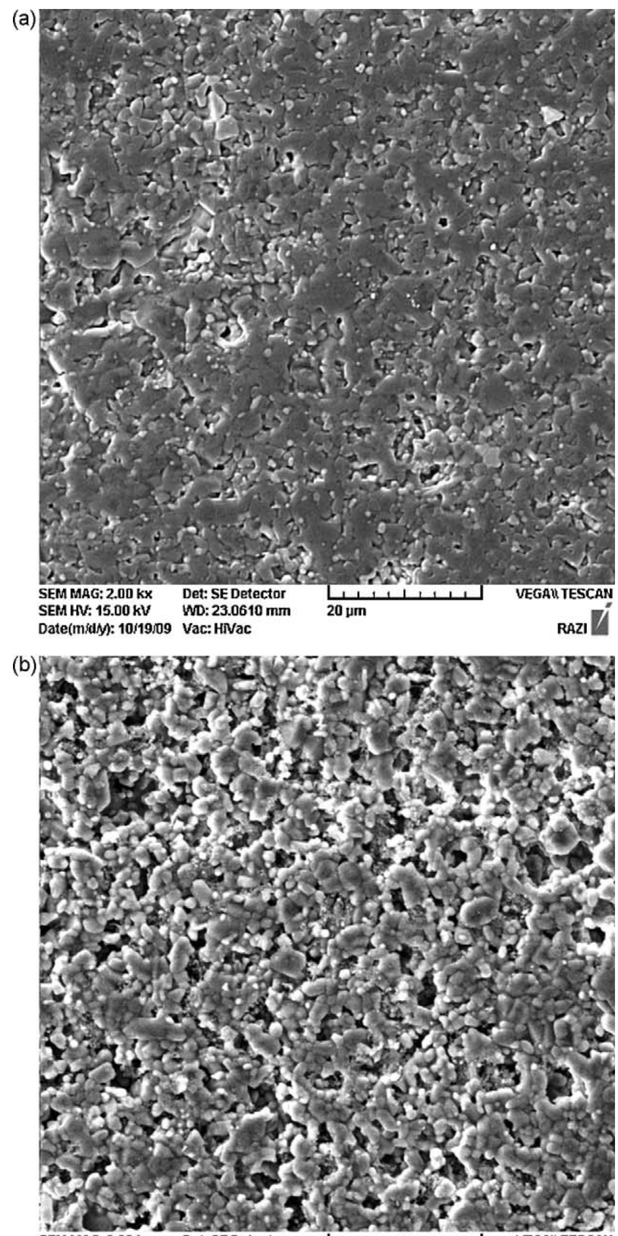


Fig. 8. SEM images of composite samples without (a) and with 20 vol.% SiC particles (b) sintered at 1650 °C for 2 h.

## 4. Conclusions

The rheological measurements of alumina–zircon–SiC suspensions were carried out. Results showed that the high stable suspensions can be obtained at pH 9 using 0.5 wt.% dispersant. This study revealed that the zeta potential values and also adsorption of dispersant on the surfaces in alumina–zircon suspensions decreased by adding SiC particles up to 20 vol.%. By increasing the SiC content to 30 vol.%, the viscosity decreased and the adsorption of dispersant on the surfaces increased due to the sedimentation behaviour of large SiC particles. The stable suspensions containing 51 vol.% solid concentration were successfully stabilized. The adsorption curves showed that Dolapix can be used as the effective dispersant for alumina–zircon–SiC suspensions. α-alumina, mullite, monoclinic- and tetragonal-zirconia, and SiC were the only crystalline phases in composite samples sintered at 1650 °C. A porous microstructure was obtained by adding SiC particles.

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