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Effect of temperature on rheological behavior of silicon carbide aqueous suspension

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Abstract

The effect of temperature on surface charge properties and rheological behavior (like pH, viscosity and zeta potential) of an aqueous SiC suspension were studied. Temperature significantly affected the rheological behavior of the aqueous SiC suspension. The pH and zeta potential of the SiC suspension all decreased with the increasing of temperature. Steady rate sweep experiments showed an increase in viscosity with increasing temperature and the effect was enhanced with increasing solid loading. These effects are attributed to a destabilizing effect of temperature, derived from the dissolution of a thin SiO_2 layer on the SiC surface.

Keywords: A. Suspensions; D. SiC; Rheology

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1. Introduction

For many applications in ceramic processing, highly concentrated ceramic suspensions are required for the final high performance products. Increasing attention is, therefore, directed on the colloidal processing method, which may result in a more homogenous green microstructure [1,2]. In all of the processing stages preceding firing, a basic step is the control of the rheological behavior of ceramic suspensions [3]. A basic problem with which this paper is concerned is the stability of colloidal suspensions. A stable colloidal suspension of repulsive interparticle forces may consolidate into a densely packed structure, whereas an unstable one where the attractive interactions between particles dominate may lead to loosely packed structures [4].

There exist many factors that can affect the colloidal stability of ceramic powders in water such as the solid volume fraction, the ionic concentration and pH value, the contemporary presence of oxides with different surface charge properties, the nature and the amount of surface active agents and dispersant.

Temperature represents a fundamental parameter in ceramic science, normally associated with heat treatments (drying, calcination, sintering), chemical stability of the compounds, and performance under operative conditions of the ultimate products. However, temperature also affects other processing steps, such as the flow behavior of suspensions and the deposition rate in liquid-based forming techniques. Some researchers [5–11] have studied the effect of temperature on colloidal stability of aqueous alumina suspensions. Tomita et al. [5] observed that temperature strongly influences the properties of electrosterically stabilized alumina suspensions. With increasing temperature, they observed that an increase of the amount of dispersant was required for attaining a minimum viscosity. A similar trend was reported by Pagnoux et al. [8]. Binner and Murfin [7] also found that increasing temperature enhanced slip flocculation of electrostatically and electrosterically stabilized aqueous alumina suspensions. Tari et al. [10] examined the effect of temperature on surface chemistry and rheological properties of electrostatically stabilized aqueous alumina suspensions. The results showed that increasing temperature leads to a gradual diminution of alumina surface ionization, dielectric constant of the medium, and a total energy barrier for coagulation. For a given system, a change in temperature will lead to: (i) an increase in thermal

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agitation (Brownian motion) [12]; (ii) a displacement of the acid/base equilibrium between particle surface and surrounding medium [13]; and (iii) a change in the dielectric properties of the medium [14]. All these changes will have competitive effects on the interactive forces between particles in suspension.

Few previous works have been undertaken into the effect of temperature on colloidal stability of aqueous SiC suspensions. The purpose of this work is to study the effect of temperature on surface charge properties and rheological behavior (like pH, viscosity and zeta potential) of electrostatically stabilized SiC aqueous suspension.

2. Experimental procedure

β-SiC ($d_{50} = 5.73$ μm, 50% of powders are under 5.73 μm) powders, manufactured by Grinding Wheel Plant, Shenyang, China, were used in this work. SEM observation (Philips SEM 515, Philips Corp. Holland) indicated that the shape of particles was irregular polyhedron. The specific surface area of the powder was measured by a single point BET method (Masterisizer 2000, Malvern Instrument Ltd., UK) to be 1.506 m^2/g.

A series of suspensions containing a certain amount of SiC powders and deionized water were mixed. The suspensions were then blended thoroughly by ball milling for 8 h using SiC spherical grinding media.

A strain-controlled rheometer (4ARES-9a, Rheometric Scientific, USA) was used to measure the rheological properties of the SiC suspensions from 20 to 90 °C. Couette (cup diameter: 36.8 mm, bob diameter: 35.0 mm, bob length: 37.37 mm) was used for all these measurements. The samples were protected from drying by adding a thin layer of paraffin oil on top. To avoid undesired influence from different mechanical histories, fresh samples were homogenized by pre-shearing at an identical rate of 100 S^{-1} for 1 min and left standing for an additional 2 min prior to measurement. Steady rate sweep measurement was used to characterize the general flow behavior with shear rates ranging from 10^{-2} to 500 S^{-1}. Zeta potential measurements were performed on BI-ZetaPlus (Brookhaven Instruments Corp., USA), which uses the Doppler shift resulting from laser light scatter from the particles to obtain a mobility spectrum. The changes in pH were followed with a PHB-1 pH-meter (San-Xin Instrumentation Inc. Shanghai, China). The suspensions were held in a container at a certain temperature and were gently mixed during the pH measurements to prevent sedimentation.

3. Results and discussions

3.1. pH change with temperature

The pH value of a slurry is a critical parameter for its stability. The variation of the pH change for deionized water and SiC suspensions as a function of temperature is presented in Fig. 1. As shown in the curves, the pH value for SiC suspensions decreases while for water stays almost constant with the increase of temperature. These variation tendencies may be rationalized as follows: possibly the following reactions [15] happen in the suspension:

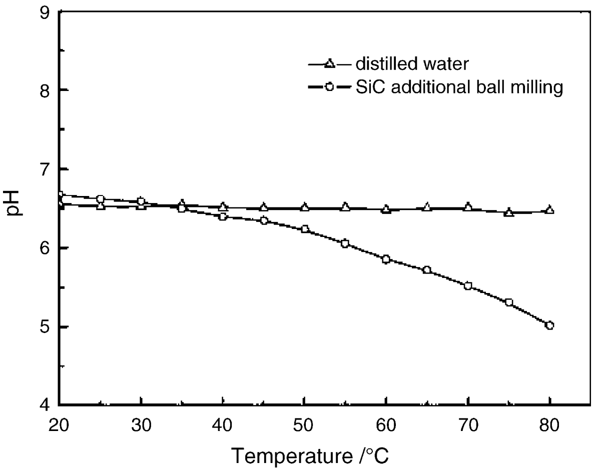

Fig. 1. pH change with temperature.

$$H_2O \Leftrightarrow H^+ + OH^- \tag{1}$$

$$Si-OH + H_2O \Leftrightarrow Si-O^- + H_3O^+ \tag{2}$$

$$SiO_2 + 2H_2O \Leftrightarrow 4Si-OH \tag{3}$$

$$SiO_2 + 2H_2O \Leftrightarrow Si(OH)_4 \Leftrightarrow SiO(OH)_3^- + H^+ \Leftrightarrow SiOO(OH)_2^{2-} + 2H^+ \tag{4}$$

Eq. (3) is a surface reaction and Eq. (2) is the dissolving of surface groups. Reactions (1) and (4) influence the pH value more than the surface reactions (2). The dissociation constant of water does not change significantly with increasing temperature (Eq. (1)). By ball milling, the SiO_2 is hydrolyzed according to Eqs. (3) and (4), which makes the solution acidic. Reaction (4) depends on the temperature; therefore, the pH value decreases with increasing temperature. The decrease of pH and the increase of ionic concentration in solution will decrease electrostatic repulsion between particles, resulting increase of flocculation trend.

3.2. Effect on zeta potential

Fig. 2 shows that the zeta potential decreases as temperature increases. This behavior can be explained by the dissolution of SiO_2 from the surface layer of the SiC powder. The solubility of SiO_2 increases with increasing temperature. The reaction (4) could happen as SiO_2 dissolves in water. The ionic concentration in the solution increases through dissolving SiO_2, consequently, the thickness of the electrical double layer and the zeta potential decreases.

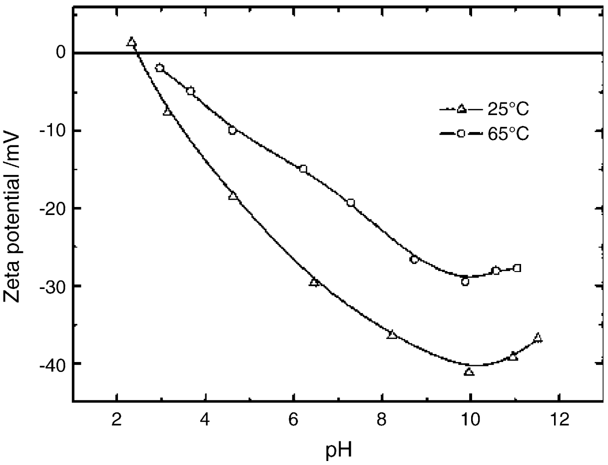

Fig. 2. Zeta potential at different temperature.

This can also be explained by the surface charge of SiO_2 (Fig. 3 [16]). The surface charge depends on the pH value. In areas of lower pH, SiO_2 gets less surface charged; therefore, a lower value of zeta potential develops. The pH value of SiC suspension decreases with raising temperature (Fig. 1); hence, the absolute value of the zeta potential decreases at elevated temperature due to the dissolution of the thin SiO_2 layer on the SiC surface, because the dissolution increases ion concentration in the suspension.

3.3. Viscosity

Although the change of pH and zeta potential show the coincident trends, they were derived from very dilute suspensions and provide little insight on the effective pair potentials in concentrated suspensions. Because of this limitation, the perceived effect of temperature was assessed by shear rate analysis of suspensions prepared at different solids loading.

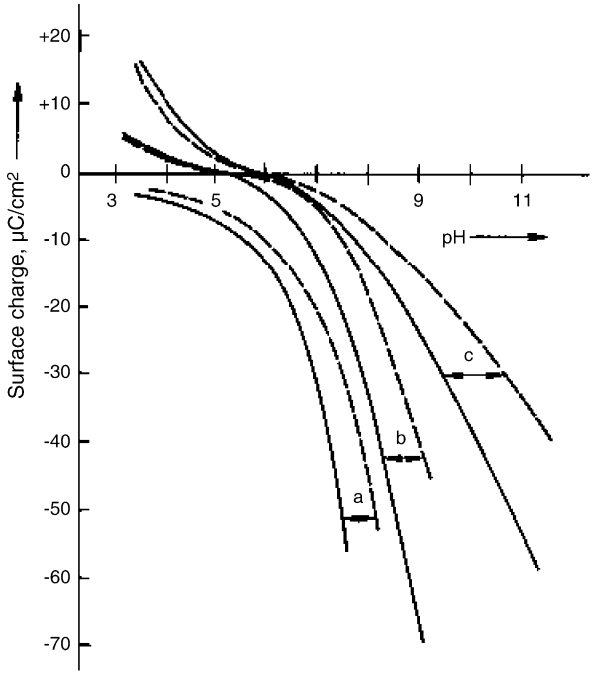

Fig. 3. Variation of surface charge of oxides with pH at 20: (a) precipitated silica; (b) TiO_2; and (c) hematite.

The influence of solid loading and temperature on the viscosity of the SiC suspensions is illustrated in Fig. 4. All of these suspensions behave as shear thinning, such shear dependence becomes much more pronounced when the temperature increased. At the solid loading of 30 vol.%, the suspensions tend to attain the Newtonian plateau at the shear rate of about 100 S^{-1} at 20 °C.

The shear-thinning behavior is usually associated with the slurry structure. At low shear rates, liquid is immobilized in void spaces within flocs, and the floc network with the surface forces dominating the particulate system. As the shear rate increases, the flocs and floc network break down, the entrapped liquid is released and a more ordered structure in the flow direction is formed. Therefore, the more pronounced shear-thinning character at high temperature can be attributed to a change in the state of dispersion towards flocculation, confirming the destabilizing effect of temperature [17].

A typical trend of viscosity versus temperature for the same slurry at different shear rates has been depicted in Fig. 5. It is shown that the viscosity rapidly increases with the increasing of temperature at low shear rate and with solids loading increase. To understand the behavior of the slurry with temperature, we have to consider the following points. Firstly, the effect of temperature on the viscosity (η) of the solvent usually follows an Arrhenius type of relationship [18] given by:

$$\eta = A e^{-B/T}$$

where A and B are constants and T is the temperature.

Secondly, the flow properties of suspensions depend upon the degree of agglomeration as a result of the dynamic equilibrium between the hydrodynamic force and the interactive force between the particles. The maximum hydrodynamic force (F_H) [11] between two touching spheres can be described as

$$F_H = 6.12\pi\eta R^2\gamma$$

where η is the solvent viscosity, R is the radius of flow unit and γ is the shear rate. It has been observed that at a given shear rate, an increase in temperature results [11] in the reduction of the hydrodynamic force (arising from the shear field), which, in turn, could stabilize more agglomerates.

And thirdly, it is known that [19] osmotic pressure differences exist between the bulk solution and the solution in the gap between the particles. Interestingly, osmotic pressure (Π) is directly proportional to temperature as given below:

$$\Pi = MRT$$

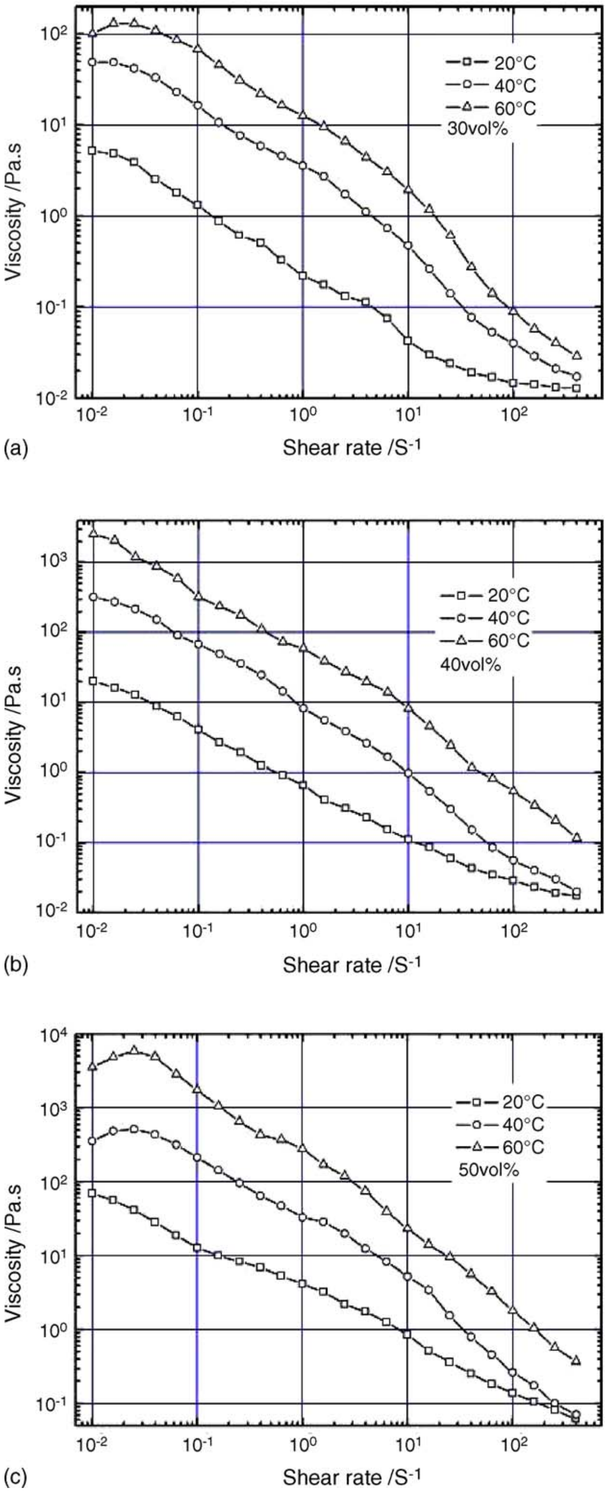

Fig. 4. Viscosity change of SiC suspension at different temperatures: (a) solid loading 30 vol.%; (b) solid loading 40 vol.%; and (c) solid loading 50 vol.%.

where M is the concentration of the solution, R is the gas constant and T is the temperature. Hence, a higher osmotic pressure at elevated temperature may enhance depletion stabilization. The interplay of all the above factors dictates the viscosity versus temperature behavior of slurry.

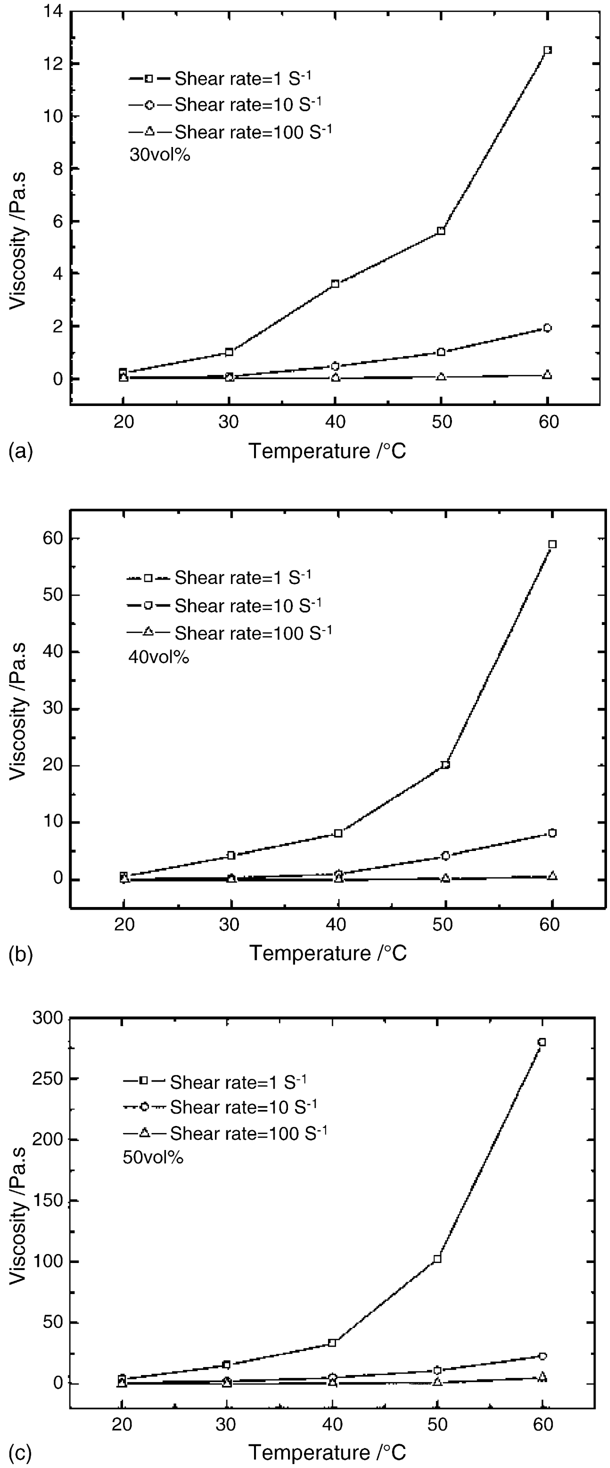

Fig. 5. Viscosity change with temperature of SiC suspension at different shear rates.

In our case, two physical parameters, viscosity and density of the liquid medium are affected by temperature. The viscosity and density of the liquid medium will mostly affect the Stokes viscous forces and gravitational/inertial forces, respectively. However, these forces are negligible when shearing or other external forces are applied to the system [10]. The variation of viscosity of water in the temperature range of −5–60 °C is less than 1 mPa S [20]. Therefore, viscosity and density variations of the liquid medium with temperature can be ignored for the present discussion. As discussed earlier, the increasing of temperature decrease the pH value of the slurry and increase the ionic concentration of the solution, which decrease the repulsion between particles. At low shear rates, the Van der walls attraction dominate the interactions of particles, SiC particles in slurry will agglomerate together. An increase in temperature may lead to increase of slurry viscosity due to the formation of more agglomerates. However, at high shear rates, agglomerates break down resulting in small change of the slurry viscosity with temperature.

4. Conclusions

The effect of temperature on aqueous SiC suspension was studied in the temperature range of 20–60 °C. The results presented in this work reveal that an increase in temperature of SiC aqueous suspensions induces a flocculation trend that is enhanced with increasing solid loading.

The pH value of SiC suspension decreases with increasing of temperature, and the zeta potential of SiC particle has the same trend. The absolute value of the zeta potential decreases at elevated temperatures due to the dissolution of a thin SiO_2 layer on the SiC surface.

Shear rate analysis confirmed the destabilizing effect of temperature. At low shear rates, the viscosity has a sharp increase due to the formation of more agglomerates. However, at high shear rates, agglomerates break down resulting in a small change of the slurry viscosity with temperature.

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