Study on Film Formation Properties of SiC Slurry

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Abstract. Two kinds of silicon carbide (SiC) powders with median diameters of 0.60μm and 2.04μm were selected as raw materials, and uniformly dispersed SiC coating film solutions were prepared through different particle grading. The effects of gradation ratio and dispersant tetramethylammonium hydroxide (TMAH) on the dispersibility and rheology of SiC coating films were studied, and the effect of dispersant content on the film forming properties of the slurry was discussed. When the fine powder and coarse powder grading mass ratio is 3:7, the particle size distribution of the mixed powder is closest to the theoretical distribution of the Dinger model. At this time, the slurry has good rheology and high particle bulk density. When 0.4% TMAH is added, the viscosity of the slurry is the lowest. When TMAH is added in an amount of 0.1 to 0.2%, the slurry has good film-forming properties.

1. Introduction
In recent decades, ceramic membranes have been successfully used in various industries such as wastewater treatment, oil-water separation, food production and gas separation. With the rapid increase of production requirements, much work has been focused on the development of ceramic membranes [1]. The silicon carbide ceramic film includes two parts, a support body and a film layer. The support body plays a supporting role as a skeleton, the membrane layer functions as a separator [2]. When coating the porous ceramic support by dipping and pulling, the thickness and integrity of the separation layer are greatly affected by the properties of the coating liquid.

In this paper, the film is coated by dripping and pulling. To study the effect of particle gradation, pH value and dispersant content on the dispersion properties of SiC coating liquid. The influence of coating film viscosity on the integrity and thickness of silicon carbide film was discussed. Full uniform film of silicon carbide ceramic membrane prepared.

2. Experimental

2.1. Preparation
The two kinds of silicon carbide powder used in this experiment were produced by Hangrui Company of Henan Yakun Group. Purity ≥ 99%, TMAH solution as dispersant (Sinopharm Chemical Reagent Co., Ltd. Quality score is 25%); the support is produced by Hubei Dijie Membrane Technology Co., Ltd.; the water used in the experiment was deionized water.

Mix silicon carbide powder and water to prepare a slurry with a solid phase volume content of 24%. The silicon carbide powder consists of coarse-grained silicon carbide SiC6000# ($D_{50}=2.0μm$) powder.
and fine-grained silicon carbide SiC10000# (D$_{50}$=0.60μm) according to the mass ratio of 1:9, 2:8, 3:7 and 4:6. TMAH was added to disperse the silicon carbide, and adjust pH by adding 0.1 mol/L HCl or NaOH, ball milling was performed at a speed of 160 r/min for 3h to obtain a SiC slurry. The slurry was coated on a silicon carbide support by a dip coating method, and was naturally dried at room temperature for 12 hours, and then dried at 100°C for 6 hours in a blast drying box to obtain a dried ceramic film blank.

2.2. Characterization

Malvern nano-particle size potentiometer (model Zetasizer Nano ZSE, Malvern,UK) was used to measure the particle size distribution of the SiC slurry and Zeta potential of the particle surface. The pH value of the slurry was measured using a Rayleigh pH meter (Shanghai Leizhou Instrument Co.,Ltd.). The viscosity of the slurry is measured by a rheometer (model RSTCC, Brookfield,USA). The shear rate ranges from 0 to 500S$^{-1}$.

3. Results and discussion

3.1. Effect of particle gradation on SiC slurry viscosity

Particle size distribution is an important factor affecting the spatial close packing of particles. Generally, a slurry prepared from a powder having a wide particle size distribution has a lower viscosity than a slurry prepared from a powder having a narrow particle size distribution. Funk and Dinger proposed the ideal particle distribution function:

$$U(D) = \frac{D^n - D_{min}^n}{D_{max}^n - D_{min}^n}$$  (1)

Among them: U(D) is the cumulative fraction of particles smaller than this size; $D_{min}$ is the smallest particle size in the powder; $D_{max}$ is the largest particle size; D is a certain particle size. The Dinger model gives an n value of 0.37. When the maximum bulk density can be reached. Figure 1 shows the particle size distribution of the SiC powder raw material.

![Figure 1](image)

Figure 1. 6000# and 10000# silicon carbide powder particle size distribution.

After grading two kinds of powders with different average particle diameters according to different mass ratios, and then milling the slurry for 3 hours, the particle size distribution was measured by a laser particle size analyzer. The data indicated by the black solid points in Figure 2 are the ideal particle size distributions calculated according to the Dinger model, and the parameters are: The minimum particle diameter of fine powder $D_{min}$=0.14μm; the maximum particle diameter of coarse powder $D_{max}$=7.45μm; n=0.37. After mixing the two powder gradings, on the whole, when the grading
mass ratio of fine powder and coarse powder is 3:7, the particle size distribution is closest to the value calculated by the Dinger model. According to the Dinger model, at this time, the powder particles can form the most efficient stacking structure, and contain the most solid phase particles in a certain space.

![Figure 2. Particle size distribution of fine powder and coarse powder according to different proportions](image)

Figure 2. Particle size distribution of fine powder and coarse powder according to different proportions

Figure 3 is a rheological curve of fine powder and coarse powder grading at different mass ratios to prepare a slurry with a solid phase volume content of 24%. According to the Dinger model, after gradation, small-sized powder particles are filled into the large-sized particle gaps, and smaller particles can replace the space occupied by the solution between large particles. The liquid medium in the original larger-sized particle gap is released, thereby increasing the content of the free solution in the slurry and improving the fluidity of the slurry. Comparing the different gradation ratios of the two powders, when the mass ratio of fine powder and coarse powder is 3:7, the slurry has the lowest viscosity and the best fluidity. This is consistent with the optimal grading ratio of the two powders obtained when the powder particles reach the closest packing from the Dinger model. Continue to increase the proportion of fine powder, but the viscosity of the slurry increases. This is because the specific surface area of the fine powder is large and the surface activity is high. The electrostatic force, van der Waals force, and capillary adsorption between the fine powder particles are relatively strong, so the slurry viscosity becomes larger. When the mass ratio of the fine powder to the coarse powder is 3:7, a slurry with high particle bulk density and good fluidity can be prepared.

![Figure 3. Rheology of SiC powder slurry with different grades](image)

Figure 3. Rheology of SiC powder slurry with different grades
3.2. Effect of Dispersant Concentration on Rheological Properties of SiC Slurry

In the preparation of the coating liquid, the addition of TMAH can not only improve the surface charge characteristics of the SiC powder, make the SiC powder uniformly dispersed in the slurry, but also have a greater impact on the rheology of the SiC slurry [3]. Figure 4 shows the effect of TMAH addition on slurry viscosity when the gradation ratio is 3:7. For a SiC slurry with a solid volume fraction of 24%, the viscosity of the slurry is higher when no dispersant is added. As the shear rate increases, the viscosity gradually decreases, which shows a tendency of shear thinning. This is a typical pseudoplastic fluid characteristic, which indicates that the dispersion of silicon carbide particles in the slurry is not good enough and is greatly affected by the external force. At low shear rates, the thermal motion of the particles exceeds the shear stress and becomes the dominant factor. The liquid medium in the space between the particles cannot flow freely. To make the total flow occur, the particles must flow around, or "bounce" the slurry between each other, due to the large resistance in this process, the viscosity is high. When the shear rate increases, the original state of the system is broken, the liquid-phase medium surrounded by the particles is released, and the applied rate gradient makes the particle structure oriented. This orientation allows the particles to pass more freely between each other, so the viscosity decreases.

For the SiC slurry system added with TMAH, as the shear rate increases, the viscosity of the slurry does not change much, which is a typical feature of Newtonian fluids. After adding an appropriate amount of TMAH, TMAH has effectively stabilized the dispersion of the slurry through the electrostatic stabilization mechanism, so the stability and rheology of the silicon carbide powder suspension are better [4]. The weak external force can hardly change the original stable equilibrium state of the system, showing the rheological behavior of near Newtonian fluid.

![Figure 4](image)

**Figure 4.** Effect of TMAH addition on slurry rheology

With the addition of dispersant TMAH, the viscosity of the slurry gradually decreased. When the mass fraction was added to 0.4%, the viscosity of the slurry reached a minimum of 4.6 mPa·S. Continue to add TMAH, the concentration of anti-particles in the solution increases and the thickness of the compressed electric double layer increases the viscosity of the slurry.

3.3. Effect of coating film viscosity on film thickness

Figure 5 shows the coating effect of a single coating film when the amount of TMAH is changed when the solid phase volume content of the slurry is 24% and the gradation ratio is 3:7. It can be seen from the figure that when TMAH is not added, the viscosity of the slurry is relatively large, the film layer obtained is too thick, and it is easy to crack during the drying process. When 0.4% dispersant is added and the viscosity of the slurry is low, the film layer made by one coating film is incomplete and the support is exposed. Therefore, when TMAH is added in an amount of 0.1 to 0.2%, a complete,
non-defective film layer can be formed by one coating.

Figure 5. Different amounts added TMAH coating results (a) 0% TMAH, (b) 0.1% TMAH, (c) 0.2% TMAH, (d) 0.4% TMAH

4. Conclusion

(1) Gradation of particles with different particle sizes, so that small particles are filled in the gaps between large particles, thereby increasing the particle packing density and reducing slurry viscosity. When the mass ratio of 10000# powder and 6000# powder is 3:7, the particle size distribution of the mixed powder is closest to the theoretical distribution of the Dinger model. At this time, the slurry rheology is good.

(2) Adding dispersant TMAH can effectively reduce the viscosity of the slurry and improve the rheology of the coating film. When 0.4% TMAH is added, the viscosity of the slurry is the lowest, but too low viscosity makes it difficult to coat the film at one time. When 0.1~0.2% TMAH is added, a complete and uniform silicon carbide film can be successfully obtained by one coating.

5. Acknowledgments

This work was financially supported by Graduate Innovative Fund of Wuhan Institute of Technology (No.CX2018056) and Project of Technology Innovation in Hubei Province (No.2016ACA161).

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