O U R N A L O F

Ceramic Processing Research

Influence of raw material grain composition on the properties of fused silica ceramics

Qingtao Wang^a, Sen Li^a, Huaqin Yu^a, Fengzhi Li^b, Huijun Xu^c, Haibo Qiao^b, Juncheng Liu^a and Qingyang Du^{a,*}

^aSchool of Materials Science and Engineering, Shandong University of Technology, Zibo 255049, China ^bZibo New Space Ceramics Limited Company, Zibo, Shandong 255190, China ^cSchool of Chemistry and Chemical Engineering, Shandong University of Technology, Zibo 255049, China

Fused silica ceramics were prepared by slip casting method with various particle sizes ($d_{50} \approx 2.31 \,\mu$ m, 19.79 μ m, and 71.46 μ m) from three types of fused silica powder. The influence of raw material grain composition on the viscosity of slurry, bulk density, porosity, water absorption, and flexural strength of the fused silica ceramics was investigated. The slurry viscosity was measured by an XND-1 viscometer. The bulk density, porosity, and water absorption of the sintered samples were obtained by the Archimedes method. The flexural strength was measured by a three-point bending test. The phase composition and microstructure of the samples were characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM). The results show that when the grain composition was 5:3:2, slurry viscosity was 150.79 mPa·s with solid content of 75%, the fused silica ceramic particles were packed most tightly and demonstrate bulk density of 2.02 g/cm³, porosity of 6.99%, water absorption of 3.47%, and flexural strength of 51.27 MPa.

Key words: Fused silica ceramics, Grain composition, Slip casting, Bulk density, Flexural strength.

Introduction

Fused silica ceramics are widely used in aviation, electronics, metallurgy, glass, etc., owing to their high thermal stability, low thermal expansion, and low thermal conductivity [1-3]. Forming methods for fused silica ceramics include slip casting, gel-casting, isostatic pressing, centrifugal casting, etc. The gelcasting method employs a large amount of organic reagents, and the density of the product is often less than 1.95 g/cm³ [4]. The isostatic pressing approach is unsuitable for large-size and complicated shape products with harsh requirement for equipments [5]. On the contrary, the slip casting method benefits from simple operation, low cost, and capability for large and complex products. Thus, it has been widely adopted in fused silica ceramic industry [6-8]. The properties of the fused silica ceramics prepared via the slip casting technique are affected greatly by the characteristics of the slurry. The size of the raw material particles has significant influence on the viscosity and dispersibility of the slurry, the bulk density, and microstructure of the samples [9]. Therefore, it is crucial to improve the properties of the fused silica ceramics by choosing the appropriate grain composition for fabricating dense samples [10].

Studies of particle packing have been conducted for many years [11, 12]. In an early study, researchers used

computer simulation method to investigate the effect of particle size distribution on the packing arrangements [13, 14]. The recent development in ceramic powder science has renewed interest in examining particle packing behavior [15, 16]. S.M. Olhero investigated the effect of particle size and particle size distribution on the rheological behavior and particle packing of silica suspension. The results showed that the viscosity of suspension increased with the addition of fine particles [17]. Wei Hu increased the bulk density of the sample up to 1.90 g/cm^3 by reducing the particle size of fused silica powder. However, the sample crystallized partially under the sintering temperature of 1300 °C [18]. Rui Wang prepared the samples with density of 1.95 g/cm³ by grain composition of two fused silica powders [19]. In order to prevent the crystallization of fused silica ceramics, it is necessary to sinter the samples at a lower temperature, so the density and the strength of the sample mainly depends on the close-packed raw materials. In our work, to obtain high density and flexural strength samples, fused silica ceramics were prepared with five different grain compositions comprising three kinds of fused silica powers. The effect of grain composition on the viscosity of slurry, bulk density, porosity, water absorption, flexural strength, phase composition, and microstructure of the samples were investigated.

Experimental Procedures

The fused silica powder was purchased from Jiangsu Zhongsheng Silicon Material Science and Technology

^{*}Corresponding author:

Tel : +86-533-2781370

E-mail: qydu@sdut.edu.cn

| Content/% 99.95 0.021 0.002 0.0013 0.0011 0.0061 0.0053 0.0041 0.01 100.00 | Composition | SiO ₂ | Al ₂ O ₃ | Li ₂ O | CaO | MgO | Na ₂ O | K ₂ O | Fe ₂ O ₃ | Ignite loss | Total |
|--|-------------|------------------|--------------------------------|-------------------|--------|--------|-------------------|------------------|--------------------------------|-------------|--------|
| | Content/% | 99.95 | 0.021 | 0.002 | 0.0013 | 0.0011 | 0.0061 | 0.0053 | 0.0041 | 0.01 | 100.00 |

 Table 1. Chemical composition of raw materials.

| Table 2. The weight ratio of raw materials for various samples. | | | | | | | | | |
|---|------------|----|----|----|----|--|--|--|--|
| Samples | S 1 | S2 | S3 | S4 | S5 | | | | |
| power A/wt% | 70 | 60 | 50 | 40 | 30 | | | | |
| power B/wt% | 20 | 20 | 30 | 30 | 30 | | | | |
| power C/wt% | 10 | 20 | 20 | 30 | 40 | | | | |

The mainly action of many meetonicle for an

Company LTD., China, which particle size was 1-2 mm. The detailed chemical composition was shown in Table 1. The raw materials were ball milled using a planetary ball mill (QM-1 SP2) to obtain three types of powder with different particle sizes. They are referred to as powder A, powder B, and powder C. The particle size distribution was 0.29-10.34 μ m, 13.52-48.17 μ m, and 71.21-129.14 μ m, measured by a laser particle size analyzer. The median diameter was 2.31 μ m, 19.79 μ m, and 91.46 μ m, respectively.

The weight ratio of power A, power B, and power C in different samples are shown in Table 2. Powders A, B, and C were first mixed uniformly with a dispersant and distilled water to form a slurry. The slurry had a solid content of 75%. Next, the slurry was poured into a porous plaster mold, vacuum degassed, and dried at room temperature for 12 hrs. Finally, the green body was dried at 110 °C for 2 hrs in a drying oven after demoulding, followed by sintering in air at 1160 °C for another 3 hrs. The fused silica samples prepared by the different grain compositions are shown in Table 2. The samples are labeled S1, S2, S3, S4, and S5.

The slurry viscosity was measured by an XND-1 viscometer according to the national standard paint viscometer, and calculated based on the outflow time of the slurry. The bulk density, porosity, and water absorption of the sintered samples were obtained by the Archimedes method. The flexural strength was measured by a three-point bending test with a support distance of 30.0 mm and a cross-head speed of 0.5 mm/min on specimens of $3.0 \times 4.0 \times 36.0$ mm. Ten specimens were measured to obtain the average strength. X-ray diffraction (XRD) patterns of the samples were recorded using a D8 Advance diffractometer with Cuka radiation. The data collection was carried out in 2θ range of 3 to 60° , with a step size of 0.02° s⁻¹. The particle size and the morphology of the final product were obtained by a scanning electron microscopy (SEM, FEI, Sirion 200).

Results and Discussion

Viscosity of slurry

The effect of the starting material grain composition on the slurry viscosity of the fused silica is studied at a slurry solid content of 75%. The results are shown in



Fig. 1. The effect of grain composition on the viscosity of slurry.

Fig. 1. The viscosity of slurry decreases with the increase of powder C amount in the raw material, and the maximum viscosity of the slurry is 447.98 mPa·s. When the grain composition is 6:2:2 and 5:3:2, the viscosity of the slurry is lower, reaching 270.49 mPa·s and 150.79 mPa·s, respectively. When the grain composition is 4:3:3, the viscosity of the slurry is 99.55 mPa·s. As the particles size distribution continues to change, the viscosity of the slurry is slightly decreased to 76.46 mPa·s.

The viscosity of the slurry varies with the particle size distribution of the raw material. As the power C content increases, the particle size increases, and the specific surface area of the particles decreases. The free water content in the slurry increases due to reduced water consumption at the same amount of powders, thus leading to a reduced slurry viscosity. Powder A is prone to agglomeration owing to the large specific surface area of the fine particles, resulting in improved slurry viscosity. This can be explained by the classical packing theory that the space between the coarse particles and the fine particles are filled with medium particles [20, 21]. Thus, the raw material grain composition has a greater impact on the flow of the slurry. The appropriate grain composition can reduce the viscosity of the slurry and improve the solid content.

Bulk density, porosity, and water absorption the samples

Fig. 2 shows the effect of raw material grain composition on the bulk density of the fused silica ceramics. The bulk density of the fused silica ceramics increases first with the addition of powder C, followed by a decrease. It can be seen from Fig. 3, the change of sample porosity demonstrates an opposite trend in comparison with the bulk density. When the grain composition is 7:2:1, the bulk density of the sample



Fig. 2. The effect of grain composition on the bulk density of the samples.



Fig. 3. The effect of grain composition on the porosity of the samples.

is 1.93 g/cm³, and the porosity is 17.52%. When the grain composition is 3:3:4, the bulk density of the sample is 1.61 g/cm³, and the porosity increases to 24.84%. When the ratio of the mixed powder is 5:3:2, the bulk density of the sample reaches 2.02 g/cm³, and the porosity is 6.99%. Compared to other ratios, the density of this sample is the largest. Therefore, varying the ratio of the mixed powder can make the sample bulk density change significantly.

Fig. 4 shows the effect of grain composition on the water absorption of the fused silica ceramics. The water absorption of the sample decreases first and then increases with the increase of the coarse particles. The variation trend of water absorption is consistent with the changing of the porosity, because the water is mainly absorbed into the pores of the sample. The ceramics show the highest density and the lowest water absorption with grain composition of 5:3:2 (Fig. 5(a)). After sufficient mixing, the pores among the irregular-shaped particles are filled with the fine particles. The gas in the pores is replaced by the fine particles, so that the bulk density increases, and the



Fig. 4. The effect of grain composition on the water absorption of the samples.



Fig. 5. SEM images of the samples, (a) S3 (b) S5.

porosity of the fused silica ceramics decreases. When the particle size distribution is 3 : 3 : 4 (Fig. 5(b)), the coarse particle content increases, and the pores among the irregular particles are filled insufficiently, and the water absorption of the fused silica increases. However, the water absorption is still lower than that of the monodisperse system, which has a large porosity [22].

The mechanical properties of the samples

Fig. 6 shows the influence of starting material grain composition on the flexural strength of the fused silica ceramics. The flexural strength of the sample increases



Fig. 6. The effect of grain composition on flexural strength of the samples.



Fig. 7. XRD patterns of the samples.

first and then decreases with the addition of powder C in the raw material. When the ratio of particles is 7 : 2 : 1, the flexural strength of the sample is 37.46 MPa. The flexural strength of the sample changes with the grain composition significantly. The flexural strength of the sample reaches the maximum value of 51.27 MPa

when the particle composition is 5:3:2. The flexural strength of the samples decreases to 39.26 MPa when the amount of coarse particles is 40 wt%.

The size of the particles has a great influence on the flexural strength. The pores produced by the accumulation of coarse particles cannot be completely filled, and the support among the particles is weak when the amount of the fine powder is small. The gaps of the skeleton formed by the coarse particles are gradually filled by increasing the amount of fine powder, so the bulk density of the sample increases, and the mechanical performance is enhanced. However, the flexural strength decreases due to the increase of the particle spacing when the powder C content exceeds 20 wt%.

XRD analysis

XRD patterns of the samples prepared with different raw materials grain composition in air at 1160 °C for 3 hrs are showed in Fig. 7. The S3, S4, and S5 samples show obvious hump-like shape, indicating that the sample is amorphous silica. Amorphous silica has low thermal expansion and high thermal shock resistance. Thus, preparation of the fused silica ceramics should



Fig. 8. SEM images of the samples ((a) S1 (b) S2 (c) S3 (d) S4 (e) S5).

avoid the crystallization of the raw materials. When the grain composition are 7:2:1 and 6:2:2, the fused silica crystallized and the low-cristobalite appears, which has a sharp peak at $2\theta = 22^{\circ}$ and 36° , consistent with PDF card 75-0923. The existence of low-cristobalite limits the application of the fused silica ceramics, because it leads to the volume shrinkage and crack in the samples [23]. The fine particles have high sintering drive due to their large surface area, which promotes the sintering of the samples. Nevertheless, the addition of more fine particles lowers the sample performance due to the crystallization of the fused silica ceramics. When the particle ratio is 5:3:2, the samples is not crystallized, while still maintains high bulk density and flexural strength.

Microstructure

Fig. 8 is the SEM images of the samples prepared using different grain compositions. The sample with grain composition of 5:3:2 has uniform particle distribution (Fig. 8(c)). The skeleton structure formed with powder C is filled with powder A and powder B into the pores, so that the flexural strength and bulk density of the samples increases and the porosity decreases. When the sample are prepared with a mixed powder ratio of 7:2:1 (Fig. 8(a)), some large pores formed by the fine particles are sintered, leading to poor mechanical properties. The sample has high bulk density due to the particles bonding together. The fracture section shows a characteristic of brittle fracture, and the medium particles from powder B are more uniformly dispersed in the sample (Fig. 8(b)). The addition of fine particles increases the density of the sample. However, the amount of coarse particles in the sample is obviously insufficient, resulting in lower flexural strength of the samples. When the particle distribution is 3:3:4 and powder C is the dominating composition, the accumulation of the particles forms more pores (Fig. 8(e)) due to the excess coarse particles, leading to large porosity and low bulk density of the sample after sintering. To obtain the amorphous fused silica ceramics, the sample should be sintered at a lower temperature, so the density of the samples mainly depends on the raw material grain composition. Therefore, optimizing the grain composition is beneficial for improving the density and flexural strength of the fused silica ceramics.

Conclusions

The raw material grain composition has significant influence on the viscosity of slurry, bulk density, porosity, water absorption, and flexural strength of the fused silica ceramics. When the course powder in the raw materials increases, the viscosity of slurry decreases, the bulk density increases first and then decreases, the porosity and water absorption decreases first and then increases, and the flexural strength increases first and then decreases. When the grain composition is 5:3:2, slurry viscosity of 150.79 mPa·s with solid content of 75%, the fused silica ceramics demonstrate bulk density of 2.02 g/cm³, porosity of 6.99%, water absorption of 3.47%, and flexural strength of 51.27 MPa. In addition, the fused silica particles were packed tightly with no crystallization. Therefore, selecting the appropriate grain composition is beneficial for improving the properties of the fused silica ceramics.

Acknowledgments

This work was supported by SDUT&Zibo City Integration Development Project (2016ZBXC083). We would like to extend our sincere gratitude for the support.

References

- 1. X.D. Yuan, W.L. Cui, W.H. Liu, Glass 26[3] (1999) 44-46.
- 2. F. Lv, C.Z. Wu, R.L. Wang, Mod. Tech. Ceram. 3 (2005) 37-40.
- A.G. Romashin, Yu. E. Pivinskii, Refrac. Ind. Ceram. 9[9] (1968) 590-595.
- 4. H.S. Hao, W.L. Cui, F.U. Peng, L.H. Xu, S.H. Wang, L. Gong, T. Song, F. Dong, Bull. Chi. Ceram. Soc. 26[5] (2007) 1036-1039.
- 5. D.M. Chen, Bei jing: National Defense Industry Press (2011) p.25.
- 6. J.J. Li, Beijing: China Light Industry Press (2001) 15.
- J.S. Lyons, T.L. Starr, J. Am. Ceram. Soc. 77[6] (1994) 1673-1675.
- Y.E. Pivinskii, F.T. Gorobets, Refract. Ind. Ceram. 9[7] (1968) 509-516.
- 9. F.F. Lange, J. Am. Ceram. Soc. 72[1] (1989) 3-21.
- 10. W.B. Wang, Beijing: Metall. Ind. Pr. (1994) p.51.
- 11. R.K. Mcgeary, J. Am. Ceram. Soc. 44[10] (1961) 513-522.
- T.G. Owe Berg, R.L. Mcdonald, R.J. Trainor, Powder Technol. [3] (1969) 183-188.
- E. Dickinson, S.J. Milne, M. Pate1, Ind. Eng. Chem. Res. 27[10] (1988) 1941-1946.
- E. Dickinson, S.J. Milne, M. Patel, Powder Technol. 59[1] (1989) 11-24.
- 15. E. Liniger, R. Raj, J. Am. Ceram. Soc. 70[11] (1987) 843-849.
- O. Koutný, J. Kratochvíl, J. Švec, et al, Pro. Eng. 151 (2016) 198-205.
- 17. S.M. Olhero, J.M.F. Ferreira, Powder Technol. 139[1] (2004) 69-75.
- W. Hu, Master's thesis, Xi An University of Architecture and Technology, Shan Xi, China, 2011.
- 19. R. Wang, Master's thesis, South China Normal University, Guang Dong, China, 2013.
- 20. C.C. Furnas, Ind. Eng. Chem. 23[9] (1931) 1052-1058.
- 21. C.C. Furnas, U S Bureau of Mines Reports of Investigations 12 (1928) p.2894.
- 22. Q.J. Zhang, Wu Han: Wuhan University of Technology Press (1997) p.135.
- B. Zheng, J.C. Liu, H.Y. Du, Rare Met. Mater. Eng. 42[6] (2013) 370-372.