Preparation and Sintering Behavior of Mullite-Alumina Composite Ceramic via Shell Coating of Nanometer Amorphous SiO$_2$ on Al$_2$O$_3$ Powder

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ABSTRACT

Mullite-alumina composite ceramic can provide improvements in some properties against individual components. However, mullite (3Al$_2$O$_3$·2SiO$_2$) exhibits relatively poor sintering properties, which originate from low diffusion coefficient. This study has attempted to overcome the difficulties in preparation by coating alumina powders by nano-meter amorphous silica, derived from sol-gel method. It is noted that alumina particles can be homogeneously coated with silica-precursor, hydrolyzed at a pH-value of 12.6 in the solution. This amorphous silica seems to give the beneficial assistance via the so-called viscous flow. So that the densification of the alumina matrix may be achieved. Thus, it has been shown that the appropriate amount of silica content is important; namely, it must be added to the amount of greater than 13wt%. The densification of the mullite-alumina composite body with a density of 96.8% (T.D.) can be obtained by sintering at a temperature of 1600°C.

KEYWORDS: mullite-alumina, amorphous silica, sol-gel, viscous flow, sintering behavior

Introduction

Generally speaking, a composite ceramic material is considered to render certain improvements in major properties in comparison to each of its component entities. Alumina (Al$_2$O$_3$) ceramic is rigid, refractory, insulating, and acid or alkali resistant; however, poor performance may result from the large coefficient of thermal expansion and poor mechanical properties under high temperature. On the other hand mullite (3Al$_2$O$_3$·2SiO$_2$) has a relatively small coefficient of thermal expansion, providing advantages over alumina in term of thermal and creep resistance. Therefore, mullite-alumina composites are fabricated to over the shortcomings of alumina.

Mullite is known to exhibit relatively poor sintering properties. Many studies[1-3] have shown that even if we adopt mullite-alumina composites using alumina powders as matrix, the large coefficient of thermal expansion and poor mechanical properties under high temperature[4] is required to give a compact sinter. Kanzaki et al.[3] pointed out that although the stoichiometry of the originating material in mole Al$_2$O$_3$/SiO$_2$ is 3/2, a small quantity of a liquid phase is needed for a densification of the sinter. His study also showed that if Al$_2$O$_3$ content is less than 74wt%, the viable liquid phase may occur at the crystal boundary and, importantly, can tend to accelerate the sintering process. At the same time, the crystal grains in the sinter grew into a long pole. If the Al$_2$O$_3$ is less than 74wt%, the viable liquid phase may occur at the crystal boundary and, importantly, can tend to accelerate the sintering process. At the same time, the crystal grains in the sinter may grow to give acicular shape. When the Al$_2$O$_3$ content is higher than 74wt%, sintering may become difficult, however, giving equilateral grains.

In the report by Cahoon et al.[4], adding in 0.05-10 wt% SiO$_2$ may hinder the sintering of Al$_2$O$_3$ because of the low diffusion rate of SiO$_2$ in Al$_2$O$_3$. Pask et al.[5,6] used to discuss the differences in sintering behavior of α-Al$_2$O$_3$ and SiO$_2$ (amorphous, quartz and cristobalite). They also pointed out that if added SiO$_2$<50 mole%, the sintering of Al$_2$O$_3$ may be severely deterred. Especially, when the temperature is above 1450°C, the sintering of sample with low SiO$_2$ concentration may be blocked obviously because of the generation of mullite. Therefore, some studies[7-8] improve the microstructure of product by using ultrafine powders of mullite. Even then, a temperature higher than 1550°C is required for a obtaining compact sinter. Although aluminum-silica gel can be used to produce mullite ceramic under a relatively low temperature[9-12], its procedures are difficult to control and prove to be uneconomical. Sacks et al.[13] selected the alumina powders and TEOS as raw materials and utilized the method of TVS (transient viscous sintering) for compaction. Thus, dense compact mullite-alumina composites can be obtained at a relatively low temperature of 1500°C.

To find out the reason for the hindrance of Al$_2$O$_3$ sintering by amorphous SiO$_2$ and the impact of transient viscous flow on Al$_2$O$_3$ sintering, we have attempted to prepare mullite-alumina composites using alumina powders as matrix, which are appropriately coated by using nanometer amorphous SiO$_2$ particles derived from an organic precursor. A discussion of the relationship between characteristics of the different powders and sintering behavior is provided.

Experimental

2.1 Preparation of powders

The raw materials for experimental were: (a) α-Al$_2$O$_3$ (AKP-50, Sumitomo Chemical Company Ltd., Tokyo, Japan) and (b) TEOS, Tetraethyl Orthosilicate (T-5083, Ferak Laboratory Chemicals, Berlin, Germany). The α-Al$_2$O$_3$ powders were dispersed by ultrasonic stirring. The aggregate was then separated from the fluid medium by sedimentation. Figure 1 shows the procedures of the experiment. First of all, the α-Al$_2$O$_3$ powders were mixed with ethanol and TEOS in a ball milling container. The NH$_4$OH had a concentration of 25%. The NH$_4$OH was added to hydrolyze the TEOS under the condition of NH$_4$OH:TEOS:C$_2$H$_5$OH = 30:1:20 (mole ratio). NH$_4$OH had a concentration of 25% and its pH was 12.6. The hydrolyzed powders were then cleaned and filtered by deionized and distilled water to clean up the remaining NH$_4^+$ and ethanol. After drying the powders at 110°C for 24 hours. Finally, the separation of target, aggregate and remaining SiO$_2$-bearing fluid were separated by sedimentation. The Al$_2$O$_3$ powders with SiO$_2$ coating were thus obtained. After calcining the powders under 600°C for 1 hour, the SiO$_2$ content of each sample is determined by EDS, Energy Dispersive Spectrometer (Jeol, JXA-840). The SiO$_2$ content of four samples A, A4, A12 and A24 are 0, 4, 13 and