ABSTRACT

Mullite and mullite based composites have been considered for a variety of high performance applications such as high temperature structural applications, infrared windows and substrates for electronic packages. These applications require high quality, high density and fine-grained ceramics. The application of sol-gel method to mullite science and technology brings new possibilities in synthesizing high purity mullite at relatively low temperatures ranging from 850 to 1250°C, yielding mullite powder with high surface area, desired size, shape and size distribution of highly sinter-active powders. In the present investigation, a systematic study has been carried out in the preparation of phase pure mullite powder, its evaluation, sintering behavior, microstructure, mechanical properties and applications for use as a self bond to the fused mullite aggregate for refractory products.

Aluminium nitrate and ethyl silicate are used as the starting materials for the preparation of mullite gel. The dried mullite gel has been used as the precursor powder. Thermal studies such as DTA, TGA and DSC are carried out and an exothermic peak at 971°C confirms the monophasic character of mullite precursor. XRD pattern reveals the structural transformation of mullite from amorphous to orthorhombic crystalline mullite through Al-Si spinel structure. The phase development and the bonding nature of the mullite precursors calcined at different temperatures
are explained using IR spectra. The effect of milling and calcination of precursor powder on particle size and surface area of the mullite powder are studied. X-ray line broadening analyses show that the ultimate crystallite size of mullite powder ranges from 20 to 30 nm. The microstructure of the dry and wet milled powders is observed using SEM. The wet milled powder shows a fine-grained microstructure.

The effect of milling and calcination of the powder on densification have been studied. The precursor, calcined at 600°C and 10 hrs of wet milling enhances the density to 90% of T.D at 1600°C for 3 hrs. The effect of suitable sintering additives such as SrO and clay on the densification of mullite is studied. The addition of 0.5 wt% of SrO enhances the density to 98% of T.D. at 1600°C for 3 hrs and the addition of 2.0 wt% of clay enhances the density to 95 % of T.D. at 1450°C for 3 hrs. When SrO is used as the sinter-additive, very fine equiaxed and elongated grains are observed. When clay is used as the sinter-additive, coarser grains with equiaxed morphology are observed.

Hardness measurements are carried out using vicker's indentor for various loads ranging from 0.5 to 10.0 kg. The sinter-additives SrO and clay increase the hardness value from 8.5 GPa to 13.5 and 11.9 GPa respectively. The additives SrO and clay also enhance the fracture toughness of mullite from 1.5 to 2.8 and 2.4 MPa m$^{1/2}$, determined by vicker's indentation method and from 1.4 to 2.5 and 2.1 MPa m$^{1/2}$ by SENB (Single Edge Notched Beam) method respectively. The modulus of elasticity is increased from 195 GPa to 225 GPa for SrO addition and 219 GPa for clay addition. Similarly, the
flexural strength (3-point bending method) increases from 183 MPa to 350 MPa for SrO addition and 280 MPa for clay addition. The fractured surfaces are mainly transgranular throughout the whole specimen.

A comparison has been made between the characteristics of the mullite products produced by the conventional clay/alumina based bonding and sol-gel derived mullite bonding for fused mullite aggregates. The density and porosity of the clay/alumina bonded and sol-gel bonded mullite are studied. The sol-gel mullite as the permanent binder yields densification of 2.48 g/cc. The room temperature flexural strength (16.9 MPa) and compressive strength (76.9 MPa) are comparable to that of the clay/alumina bonded mullite products. However, superior high temperature mechanical properties such as Hot Mo R (4.7 MPa), the retained flexural strength of 11.7 MPa after 10 thermal cycles of ΔT = 1000°C and refractoriness under load (RUL>1800°C) are obtained for the samples of fused mullite bonded by sol-gel mullite.