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Effect of yttria on sintering and microstructural behavior of reaction sintered mullite based on bauxite, fly ash and precipitated silica

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ABSTRACT

The present investigation on the effect of Y_2O_3 towards the sintering behavior of mullite compacts revealed that rapid mullitization occurred through nucleation and normal grain growth due to the formation of yttrious silicate glassy phase. The intergranular voids were progressively eliminated by yttrious silicate glass leading to significant decrease in porosity with the corresponding remarkable rise in mechanical strength of sintered compacts. The uniform dispersion of microfine corundum grains into the mullite matrix with 1.5% Y_2O_3 content was noticed during sintering at 1550 °C and above.

1. Introduction

In recent times, mullite has been emerging as an advanced structural and functional ceramics. The technical importance of mullite ceramics is evolved from its inherent properties like low thermal conductivity, excellent creep resistance, exceptional chemical resistance and outstanding thermal stability, low thermal expansion and good mechanical strength even after exposure to the severe atmospheric condition. Low dielectric constant [1] and good transparency for the mid-infrared region [2] makes mullite to a potential substrate in hi-tech electronic and optical devices respectively. Different conventional or non-conventional techniques of mullite powder synthesis viz sintermullite, fused-mullite and chemical-mullite [3] have been successfully explored. Mullitization at relatively low temperature is accomplished using high purity precursors via sol-gel route while solid-state sintering of powdered raw materials requires temperature higher than1600°C. However, mullite derived from high-purity precursor is quite expensive and not a viable option for its large-scale production due to low yield and complex processing technique. Therefore the development of novel mullite body through effective utilization of naturally occurring abundant minerals such as kaolin [4–6], sillimanite [7–9], andalusite [10,11], natural topaz [12], clay [13] etc. has now attracted much attention of the scientists.

Fly ash, a by-product of the thermal power plant, consists of both amorphous components like SiO_2, Al_2O_3, Fe_2O_3, Fe_3O_4, TiO_2 and CaO [14] along with quartz and mullite as crystalline phases [15]. Fly ash is extensively studied for preparing mullite ceramics [16]. Different sintering aids such as TiO_2 [17], AlF_3 [18], MgO [19,20], Fe_2O_3 [21] are reported to facilitate mullite formation from fly ash at a moderate sintering temperature possessing better thermo-mechanical properties. Mullite can also be prepared from fly ash [22], clay and alumina without any sintering aid. Studies revealed that substantial mullite phases are developed by using fly ash of finer particle sizes [23]. Higher mullite content body having reasonable mechanical strength over 200 MPa is successfully derived by sintering of as-received high alumina fly ash at 1400 °C and beneficiated fly ash without incorporating additional alumina at the temperature of 1500 °C [24].

Bauxite is the most important hydrated alumina rock consisting of one or more hydrated mineral phases such as gibbsite, boehmite, bayerite and diaspore in association with certain impurities like quartz, goethite, hematite, anatase, alkali and alkaline-earth oxides [25]. The presence of impurities like Fe_2O_3 and TiO_2 cause low refractoriness under load while the presence of CaO [26] in bauxite is more detrimental due to the formation of low melting vitreous phase, thereby affecting the refractory properties of mullite. High-quality dense mullite is prepared by reaction sintering of bauxite and kaolinitic clay with an enhanced bulk density of 2.89 g/cc and porosity as low as 0.58% during firing at 1500 °C [27]. The morphology of the fired compact is consisted of two types of mullite such as elongated primary mullite and equiaxed secondary mullite.

The mullite-corundum composites [28] are derived by the reaction sintering of silica sol and bauxite. Flexural strength and elastic modulus of the sintered compact at 1600 °C are markedly improved due to increased free corundum phase within the mullite matrix. Doping of V_2O_5 assists in low temperature sintering of bauxite-fly ash mixture with good sintered properties [29]. 10 mol% V_2O_5 doped mullite ceramics

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Table 1
Chemical analysis of the raw materials (wt%).

Raw materials	Constituents							
	Al_2O_3	SiO_2	Fe_2O_3	MgO	CaO	TiO_2	Alkali (Na_2O + K_2O)	LOI
Bauxite	60.30	2.90	1.85	–	1.69	0.51	0.20	32.55
Fly Ash	29.61	63.45	2.50	2.05	1.41	–	–	0.99
Precipitated Silica	–	86.55	–	–	–	–	Trace	13.40

fired at 1500 °C has been reported to exhibit substantial bending strength and low water absorption along with uniform microstructure of cuboidal shaped mullite grains.

In the present study, a comprehensive attempt has been made to evolve dense mullite compact by reaction sintering of refractory grade bauxite, fly ash and precipitated silica. The effect of Y_2O_3 as a dopant on the sintering temperature, phase assemblage, microstructure evolution and thermo-mechanical properties of such mullite compacts are systematically analyzed.

2. Material and methods

Refractory grade bauxite (Saurashtra, Gujarat), fly ash (Bakreswar Thermal Power Plant, West Bengal) and precipitated silica (synthesized) were taken as starting materials. In order to maintain the exact molar ratio of Al_2O_3/ SiO_2 as 3:2 in mullite precursor, the requisite amount of bauxite, fly ash and precipitated silica were chosen. Based on the chemical analysis of the starting materials, batch composition and oxide equivalent of stoichiometric mullite were computed. Table 1 summarized the chemical composition of bauxite, fly ash and precipitated silica in wt% respectively. The bauxite exhibited a high content of alumina with low percentages of associated impurities. Fly ash analysis revealed the presence of high silica and alumina content with the controlled amount of iron oxide, lime and magnesia. The chemical analysis results of starting materials confirmed that impurities were within a reasonable limit and did not grossly affect the different properties of fired mullite compacts.

Bauxite was, at first, crushed, ground and subsequently passed through BS 100 mesh sieve. The powder raw materials in requisite proportion for mullite were wet milled in a rapid pot mill using alumina balls for 10 h. The slurry thus obtained was thoroughly dried at 110 °C, calcined at 600 °C for 2 h followed by grinding of the agglomerate to a fine powder and finally sieved through BS 150 mesh.

The different batches, as given in Table 2, were prepared by intimate mixing of above stoichiometric precursor of mullite through the incorporation of Y_2O_3 in the proportion of 0, 0.2, 0.4, 0.8, 1.2, 1.5% (w/w) respectively in a high-speed planetary mixer. Rectangular bars (72 mm × 11 mm × 8 mm) were fabricated by uni-axial pressing from respective batch powder in a hydraulic press at the pressure of 150 MPa. Finally, the dried bars of each set were fired at 1400 °C, 1450 °C, 1500 °C, 1550 °C and 1575 °C respectively in a programmable muffle furnace with fixed 4 h soaking time.

Table 2
Batch compositions of mullite precursor (wt%).

Raw materials	Batch composition					
	1	2A	2B	2C	2D	2E
Refractory grade Bauxite	75.50	75.35	75.20	74.90	74.60	74.37
Fly Ash	17.50	17.46	17.43	17.36	17.28	17.23
Precipitated Silica	7.00	6.99	6.97	6.94	6.92	6.90
AR grade Y_2O_3	0	0.20	0.40	0.80	1.20	1.50

Densification behavior of the sintered samples was evaluated by measuring firing shrinkage, bulk density, apparent porosity, modulus of rupture, thermal shock resistance and residual strength. The sintered products were also tested for phase assemblage by XRD and microstructure by SEM study. Linear firing shrinkage was studied by measuring the dimensional change of the bars before and after firing (ASTM C179-14). Bulk density and apparent porosity were determined by conventional liquid displacement method using Archimedes' principle in water medium (ASTM C20-00(2015)). The flexural strength of fired compacts was measured by three-point bending method (ASTM C1161-13) using instrument LRX, version 2.13, Lloyd, UK. Thermal shock resistance was measured by placing the fired bar compacts at 1000 °C in an electric muffle furnace for 10 mins followed by placing it in the air for 10 mins to complete one cycle. After 20 cycles of spalling, the residual strength was measured by three-point bending method (ASTM C1161-13). The powder XRD patterns were recorded over Bragg's angle 2θ from 10°–60° in a Philips X-ray diffractometer (X'Pert PRO PW-3071) using Ni-filter Cu-Kα radiation at the scanning rate of 2°/min. Microstructural characterization on an etched surface of each sample sintered at different temperatures was performed by Scanning Electron Microscope (ZEISS, INCA Penta FETx3, MODEL, EDS8100, UK).

3. Results and discussion

The chemical analyses of the starting materials, as well as batch compositions used in the study, have been given in Tables 1 and 2 respectively. The oxide compositions of different mullite batch precursors are also presented in Table 3.

In each batch, a more or less constant molar ratio for Al_2O_3 and SiO_2 as 3:2 is maintained although composition contains some percentages of Fe_2O_3, CaO, TiO_2 and alkalies (Na_2O + K_2O) as impurities. Negligible reduction in the content of minor constituents occurs in oxide compositions from Batch 1 to Batch 2E.

All the samples after firing at temperatures from 1400 °C to 1575 °C are grayish-white in color and exhibit smooth texture without any distortion or crack.

3.1. Linear firing shrinkage

The variation of firing shrinkage (%) against firing temperature and Y_2O_3 content is graphically represented in Fig. 1. It is evident from the graph that with increasing temperature of firing from 1400 °C to 1575 °C, the linear shrinkage of the compacts increased continuously. The increase in linear shrinkage with the rise in firing temperature is attributed to the intense interaction of amorphous silica with transition alumina to form mullite. It is well established that cristobalite formation increases densification by intensifying mullitization process with the removal of pores and coalescence of the grains. Glassy phase formed is being progressively consumed by alumina to yield more mullite at the higher temperature of firing. In addition, mullite grains tend to grow in size. The dopant added fired samples exhibit a higher degree of firing shrinkage than that of dopant-free mullite body during firing (1450 °C to 1575 °C). Maximum firing shrinkage value of 19.23% is obtained in the mullite compact with 1.5% Y_2O_3 content during firing at 1575 °C indicating rapid material interaction, transportation and consequent removal of pores.

3.2. Bulk density & apparent porosity

The variation in bulk density vis a vis apparent porosity of sintered mullite bodies against firing temperature and dopant content is shown in Figs. 2 and 3 respectively. It is evident from the Fig. 2, that the bulk density of sintered compacts increases sharply with a simultaneous significant reduction of apparent porosity during firing from 1450 to 1575 °C. With the addition of Y_2O_3, mullite grains tend to grow large enough to come closer with adjacent grains leaving behind few voids at