

# Effect of Boehmite Addition on Sintering Behaviour of Mullite Precursor Synthesized from Bauxite, Fly Ash and Precipitated Silica

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Cost-effective mullite body was prepared by reaction sintering of natural bauxite, fly ash, a by-product in thermal power plants and precipitated silica. The study was designed to analyse the effect of synthetic boehmite as dopant on sintering behaviour of stoichiometric mullite body in terms of sintering temperature from 1400–1575 °C and dopant content varying from 0–1,2 mass-%. Sintered mullite was characterized by firing shrinkage, bulk density, apparent porosity, bending strength measurement, X-ray diffraction analysis and SEM studies. The result indicates that significant secondary mullitisation takes place at a lower temperature in presence of sintering aid. 1,2 % addition of boehmite generates remarkable thermo-mechanical properties in sintered body due to interlocked morphology with uniform distribution of the normal size mullite along with dispersion of fine corundum grains during sintering at 1575 °C.

## Introduction

Mullite is well emerged as an advanced structural and functional ceramics. Mullite finds widespread industrial applications from traditional heavy-duty refractories to optical materials as well as electronic substrates because of its superior mechanical strength, low thermal conductivity, good creep resistance, excellent chemical and thermal stability. Low dielectric constant [1] and good transparency in mid-infrared light [2] focuses mullite as a promising material in advance electronic and optical objects respectively. Different conventional or non-conventional techniques of synthesizing mullite precursor powder viz sinter-mullite, fused-mullite and chemical-mullite [3] have been extensively followed using high purity reagents/precursors. Abundantly available naturally occurring minerals and industrial waste have always been utilized judiciously for large scale and cost-effective production of mullite.

Mullite is the only crystalline phase in the binary system of silica-alumina. It can be designated by the general formula  $Al_2(Al_{2+2x}Si_{2-2x})O_{10-x}$ , where x varies from 0,17–0,50 [4–5]. High temperature mullite ( $3Al_2O_3 \cdot 2SiO_2$ ) has a density of approximately 3100 kg/m<sup>3</sup> representing the most thermodynamically stable crystalline form under high pressure and temperature [6]. Mullite material represents amorphous and crystalline structures including: spinel, tetragonal and orthorhombic. Primary mullite (MI) with cuboidal shape, is derived by decomposition of spinel-type structure of metakaolin and the residual amorphous silica reacts with excess  $Al_2O_3$  producing secondary mullite. Secondary mullite is formed from a major solution-precipitation process via a transitory liquid phase as well as a minor solid state interdiffusion due to viscosity gradient, forming two kinds of morphology; a granular crystallite size distribution denoted as MII-mullite and elongated with acicular or needle like grains namely MIII-

mullite [7]. The MIII-mullite type is formed due to the presence of dopant facilitating growth of these mullite seeds which grew as their crystallization habit (orthorhombic needles or columns). Tertiary mullite crystal is formed at the edges of corundum crystals by dissolution and further precipitation of alumina-rich glassy matrix.

Bauxite is the most important hydrated aluminous rock consisting of one or more hydrated alumina mineral phases namely gibbsite, boehmite, bayerite, diaspore and tohdite. It is however associated with some impurities like quartz, goethite, hematite, anatase along with alkali and alkaline-earth oxides [8]. The presence of impurities like  $Fe_2O_3$  and  $TiO_2$  badly affects refractoriness under load due to formation of low melting liquids. However, instead of forming vitreous phase, if  $TiO_2$  and  $Fe_2O_3$  are precipitated out of solid solution and permeate to the mullite lattice by cation exsolution mechanism [9], it may play a positive role in im-

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**Tab. 1 Chemical analysis of the raw materials [mass-%]**

Raw Material	Constituent							
	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	MgO	CaO	TiO <sub>2</sub>	Alkali	L.o.I.
Sourashtra bauxite	60,30	2,90	1,85	–	1,69	0,51	0,20	32,55
Bakreswar fly ash	29,61	63,45	2,50	2,05	1,41	–	–	0,99
Precipitated silica	–	86,55	–	–	–	–	Trace	13,40

**Tab. 2 Batch compositions of mullite precursor [mass-%]**

Raw Material	Batch Composition				
	1	2A	2B	2C	2D
Sourashtra bauxite	76,72	76,57	76,41	76,11	75,81
Bakreswar fly ash	16,78	16,75	16,71	16,65	16,58
Precipitated silica	6,50	6,49	6,47	6,45	6,42
Synthetic boehmite	0,00	0,20	0,40	0,80	1,20

proving the physico-mechanical properties of reaction sintered mullite.

On the other hand, fly ash, a byproduct of thermal power plant, consists of both amorphous materials like SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, Fe<sub>3</sub>O<sub>4</sub>, TiO<sub>2</sub> and CaO [10] and crystalline components like quartz and mullite [11] as well permitting its various applications.

A great deal of investigations has been carried out to study the role of sintering aids/dopants on sintering temperature, crystalline phase formation, morphological changes, physical and mechanical properties of mullite. It has been accepted that mullite formation at earlier stage reduces the densification process while densification prior to mullitisation is a desirable phenomenon for achieving better properties [12]. Therefore, a comparatively less reactive alumina-silica rich mixture is likely to exhibit better densification and the presence of sintering aids may promote mullitisation through liquid phase formation even at moderate sintering temperature. Y<sub>2</sub>O<sub>3</sub> [13] assists in densification by liquid phase sintering mechanism with formation of secondary mullite whereas La<sub>2</sub>O<sub>3</sub> [14] addition shows effective densification at 1500 °C with good mechanical properties due to development of anisotropic growth of mullite grains and generation of interlocking network morphology. Optimum amount of CeO<sub>2</sub> [15–16] addition favours densification and lowers the mullitisation temperature. Addition of CeO<sub>2</sub> beyond the critical limit favours formation of large amount of low viscosity CeO<sub>2</sub> – Al<sub>2</sub>O<sub>3</sub> – SiO<sub>2</sub> system which causes pore closure and subsequently reduces

the diffusion of oxygen into the matrix and thereby inhibits mullitisation. B<sub>2</sub>O<sub>3</sub> [17] reacts with alumina forming aluminium borate, which acts as seed for crystallization of mullite. Considerable amount of mullite formation along with good mechanical properties are achieved by adding CoO [18] additive in Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> system. Small amount of Gd<sub>2</sub>O<sub>3</sub> [19] doping exhibits decrease in mullitisation temperature. Fluorides (CaF<sub>2</sub>, NaF, KF etc.) shows good sinterability at lower temperature due to liquid phase sintering whereas at higher temperature extra amount of fluoride affects the properties of mullite owing to the formation of sealed porosity and glassy phase [20]. Additives like TiO<sub>2</sub> or V<sub>2</sub>O<sub>5</sub> [21] positively influences the densification process and the sintering mechanism of mullite, however, TiO<sub>2</sub> affects the high temperature bending strength [22]. Aluminium powder [23] favours columnar mullite because of high nucleation rate which facilitates growth of mullite from the wetting interface due to presence of molten alumina acting as a nucleating agent. Presence of synthetic boehmite [24] or MgO [25] increases the grain growth and subsequently increases in density. Both MgO and CaO [26] are reported to reduce the mullitisation temperature. Complicated reaction sequences are observed with pentoxides like Nb<sub>2</sub>O<sub>5</sub> or Ta<sub>2</sub>O<sub>5</sub> [27]. In presence of Nb<sub>2</sub>O<sub>5</sub>, AlNbO<sub>4</sub> is formed which retards dissolution of alumina into the silica-rich liquid phase or AlNbO<sub>4</sub> consumed Al<sub>2</sub>O<sub>3</sub> before mullite formation and thereby inhibits the mullitisation process. Ta<sub>2</sub>O<sub>5</sub> reacts Al<sub>2</sub>O<sub>3</sub> to form AlTaO<sub>4</sub> and thereby restrains mullite formation.

In the present study, a novel attempt has been made to evolve in-situ mullite by reaction sintering of refractory grade bauxite, fly ash and precipitated silica. The effect of boehmite addition on the sintering temperature, phase assemblage, microstructural aspects and thermomechanical properties of mullite compacts are systematically analysed.

## Experimental

Refractory grade bauxite (Sourashtra, Gujarat/IN) and fly ash (Bakreswar Thermal Power Plant, West Bengal/IN) and synthetic precipitated silica were selected as major raw materials. In order to maintain the exact molar ratio of Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> as 3 : 2 in mullite precursor, requisite amount of bauxite, fly ash and precipitated silica were chosen. Based on the chemical analysis of the starting materials, oxide equivalent and stoichiometric composition of mullite were derived. Tab. 1 summarizes the chemical composition in weight percentages of bauxite, fly ash and precipitated silica respectively. The bauxite exhibits high content of alumina with low percentages of associated impurities. Fly ash analysis reveals the presence of high silica and alumina content with controlled amount of iron oxide, lime and magnesia. The analysis confirms that impurities are within reasonable limits and would not grossly affect the properties of fired mullite compacts.

Bauxite was crushed, ground and subsequently passed through BS 100 mesh sieve. The powder raw materials in requisite proportions were wet milled in a ball 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 the agglomerate to fine powder and finally sieved through BS 150 mesh.

The different batches, as given in Tab. 2, were prepared by mixing of above stoichiometric precursor of mullite through addition of boehmite (synthesized from aluminium isopropoxide [28]) as nucleating agent in the proportion of 0; 0,2; 0,4; 0,8; 1,2 mass-% respectively in a high speed planetary mixer. Rectangular bars (72 mm × 11 mm × 8 mm) were fabricated by uniaxial pressing of respective batch powder in a hydraulic press at the pressure of 150 MPa. Finally, the dried bars were fired at 1400 °C, 1450, 1500, 1550 and 1575 °C respectively

in a programmable muffle furnace with fixed 4 h soaking time in each case.

Densification behaviour of the sintered samples was evaluated by measuring bulk density, apparent porosity and modulus of rupture. Linear firing shrinkage was studied by measuring the dimensional change of the bars before and after firing. The sintered products were also characterized for phase assemblage and microstructure by XRD and SEM studies. Bulk density and apparent porosity were determined by conventional liquid displacement method using Archimedes' principle in water medium. The powder XRD patterns was recorded over Bragg's angle  $2\theta$  of 10–60° in a Philips X-ray diffractometer (X'Pert PRO PW-3071) using Ni-filter  $\text{Cu-K}\alpha$  radiation at a scanning rate of 2°/min. Microstructural characterization

**Tab. 3 Oxidic composition of the different batches [mass-%]**

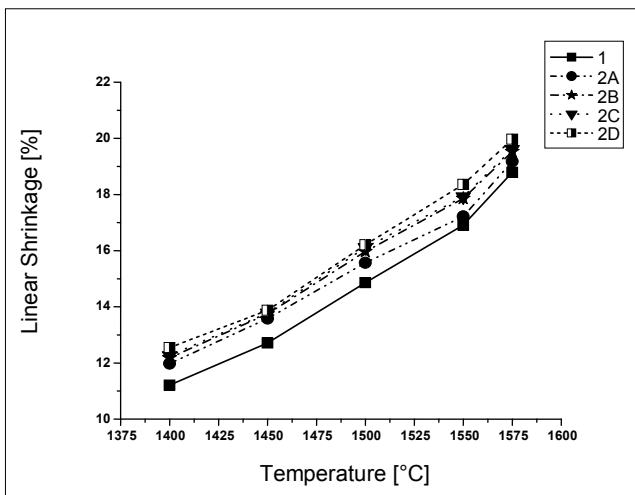
Batch	$\text{Al}_2\text{O}_3$	$\text{SiO}_2$	$\text{Fe}_2\text{O}_3$	MgO	CaO	$\text{TiO}_2$	Alkali ( $\text{Na}_2\text{O}+\text{K}_2\text{O}$ )	L.o.I. [%]
1	51,23	18,5	1,84	0,34	1,53	0,39	0,15	26,01
2A	51,33	18,46	1,84	0,34	1,53	0,39	0,15	25,96
2B	51,43	18,42	1,83	0,34	1,53	0,39	0,15	25,91
2C	51,62	18,35	1,82	0,34	1,52	0,39	0,15	25,8
2D	51,82	18,28	1,82	0,34	1,51	0,39	0,15	25,7

of etched surface of sintered samples was performed by Scanning Electron Microscopy (ZEISS, INCA Penta FETx3, MODEL, EDS8100/UK).

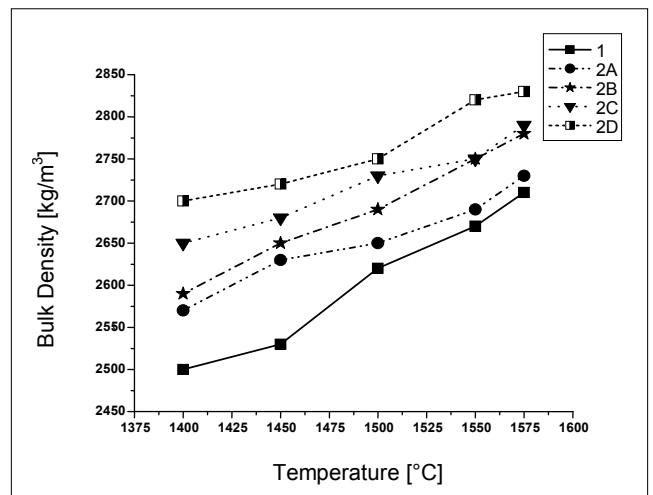
**Results and discussion**

The composition of raw materials used in preparing the mullite plays a vital role in quality and performance behaviour of the

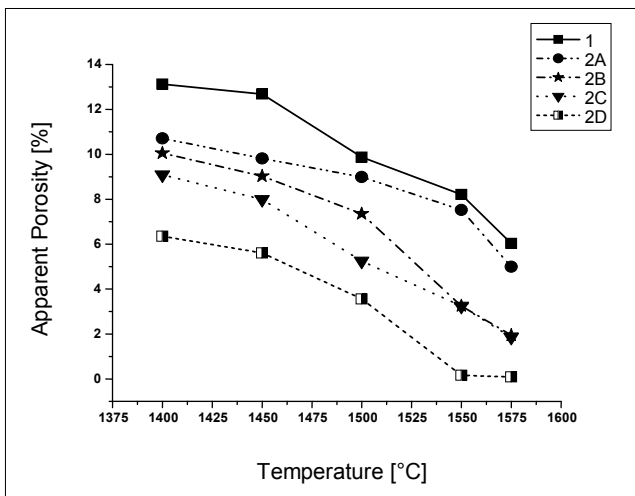
end product. The raw materials used in the study were chemically analysed and results are shown in Tab. 1. The composition of batches in terms of oxides is given in Tab. 3. The variation of firing shrinkage against firing temperature and boehmite content is graphically represented in Fig. 1. It is evident from the figure, that with increasing temperature of firing from 1400 °C to 1575 °C, the



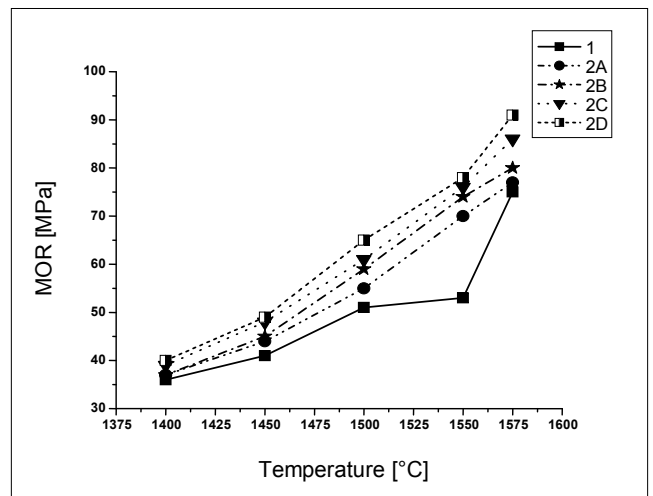
**Fig. 1** Variation of firing shrinkage of sintered compacts with temperature



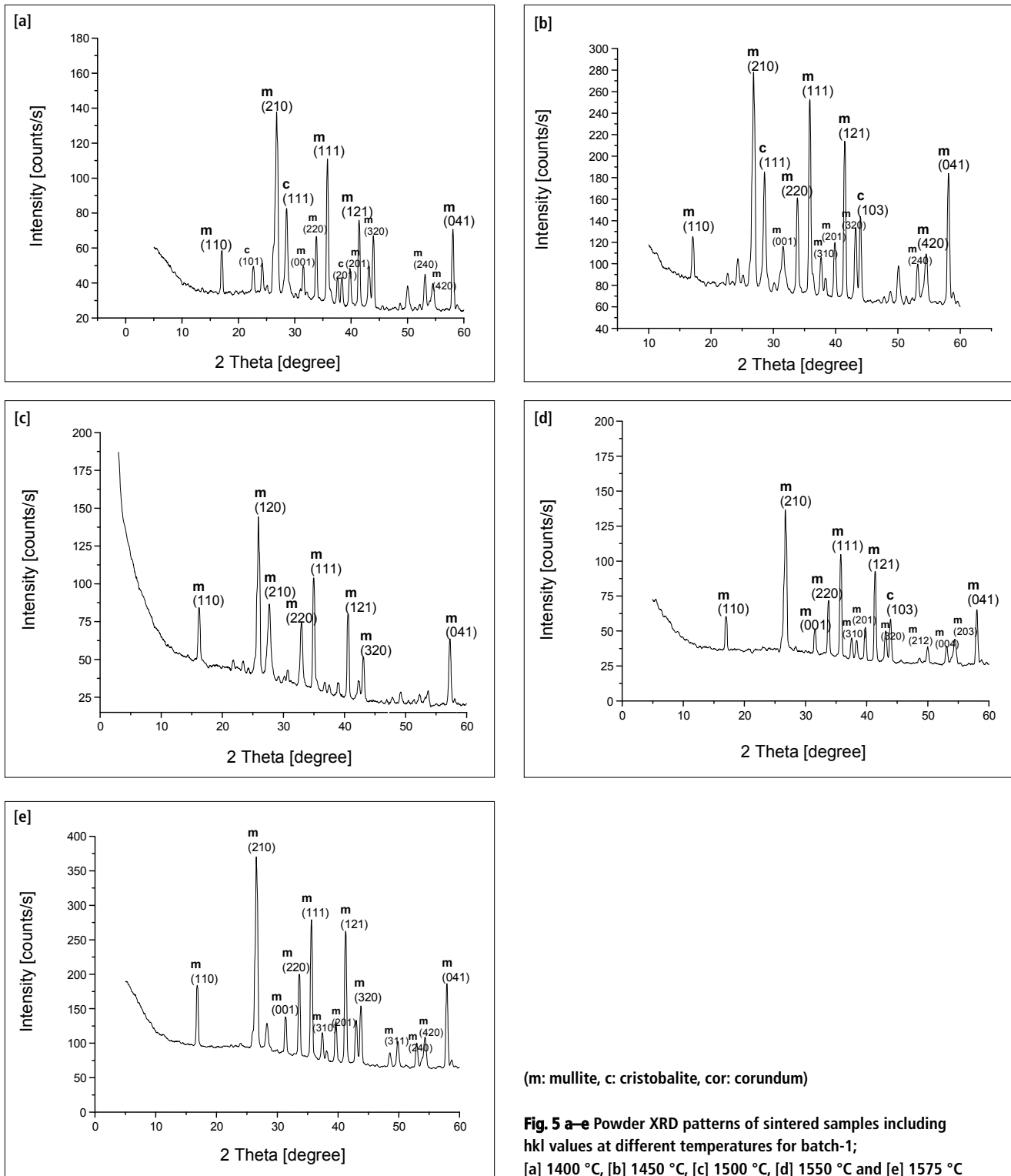
**Fig. 2** Variation of bulk density of sintered compacts with temperature



**Fig. 3** Variation of apparent porosity of sintered compacts with temperature



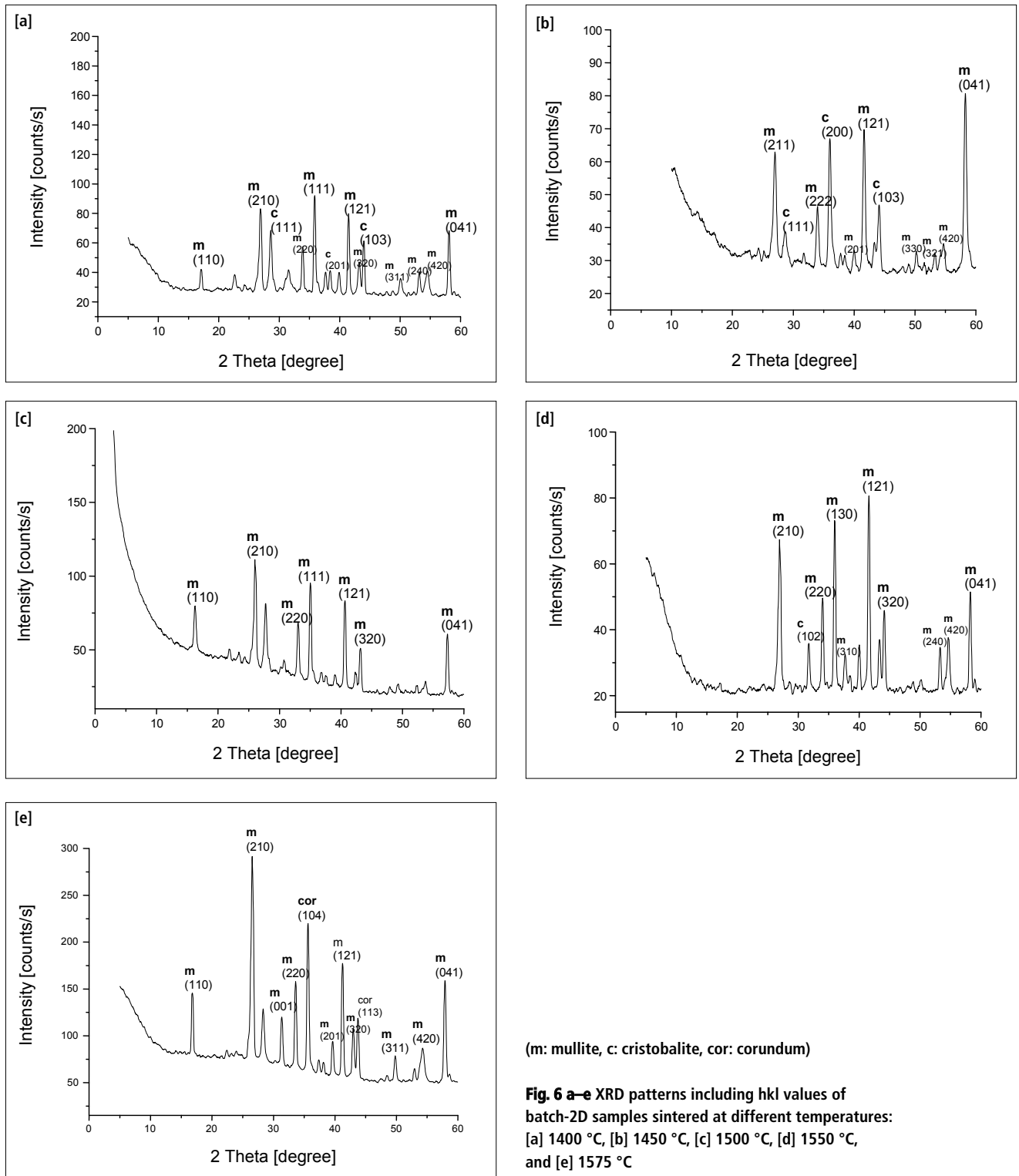
**Fig. 4** Variation of bending strength sintered compacts with temperature



linear shrinkage of the compacts increased continuously. The increase in linear shrinkage with increase in temperature is attributed to the transformation of metastable siliceous amorphous forms to crystalline cristobalite which further reacts with alumina to form mullite. It is well-established

that cristobalite formation increases densification reducing reaction paths for Al- and Si-ions respectively leading to mullitisation intensification with pores consolidation, pores removal, components shrinkage and finally grain growth. Glassy phase is consumed by alumina to form mullite at higher

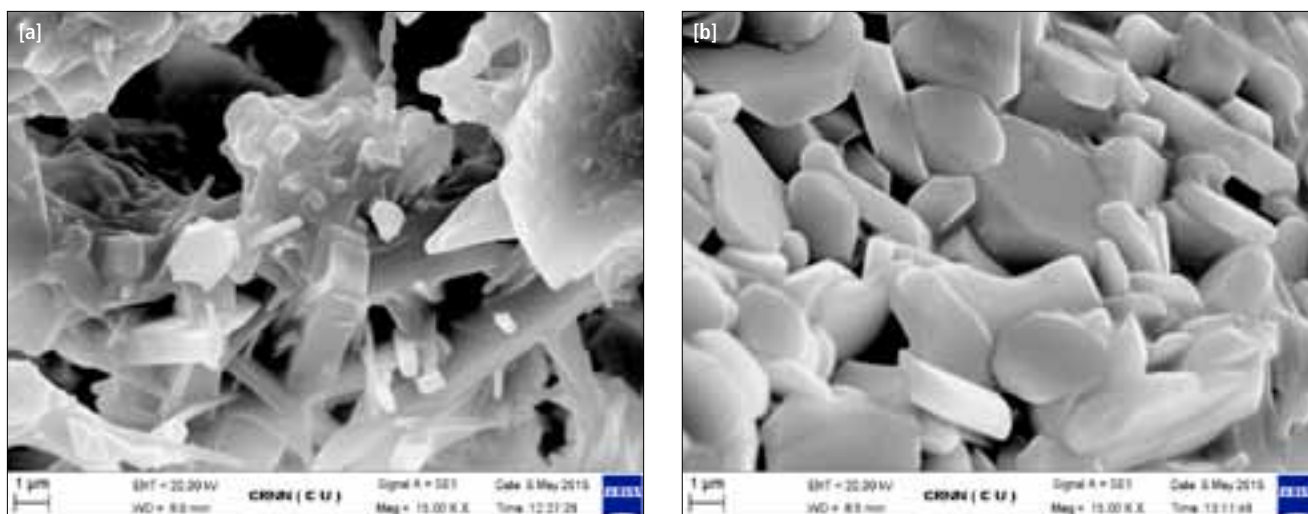
temperature; furthermore, mullite grains grow in their size. The dopant containing fired samples exhibit higher degree of shrinkage from 1450 °C to 1575 °C than that of dopant-free mullite body. Maximum firing shrinkage of 19,97 % was obtained in the mullite compact with 1,2 % addition of



boehmite precursor during firing at 1575 °C indicating enhanced elimination of pores by rapid material interaction, transportation and uniform interlocking of the grains. The variation in bulk density vis-a-vis apparent porosity results of sintered mullite bodies against firing temperature and do-

paint content are shown in Fig. 2–3. With increase in firing temperature and boehmite content in the composition, more and more densification occurs. As reflected from the Fig. 2, the BD of sintered compacts increases sharply with significant reduction of apparent porosity simultaneously during firing

from 1450 °C to 1575 °C. Active alumina in synthetic boehmite tends to react with cristobalite readily to generate mullite even at a lower temperature of 1550 °C which is responsible to yield high bulk density of 2830 kg/m<sup>3</sup> with corresponding elimination of open porosity in sintered compact.



**Fig. 7 a–b** SEM micrographs of batch-1 samples sintered at 1400 °C [a], and 1450 °C [b]

Fig. 4 reflects that boehmite addition results in high bending strength which may be due to the uniform distribution and interlocking of secondary mullite grain throughout the matrix. With increase in boehmite addition up to 1,2 %, the bending strength is improved steadily from 75–91 MPa. It is also observed that incorporation of 1,2 % boehmite precursor does not cause any cracks or deformation in the sintered mullite in the temperature range due to the higher loss of structural water from boehmite precursor itself.

Fig. 5 represents the powder XRD patterns of the sintered compacts fired at different temperatures. Batch-1 samples fired at 1400 °C exhibit mullite (PDF No 15-0776)

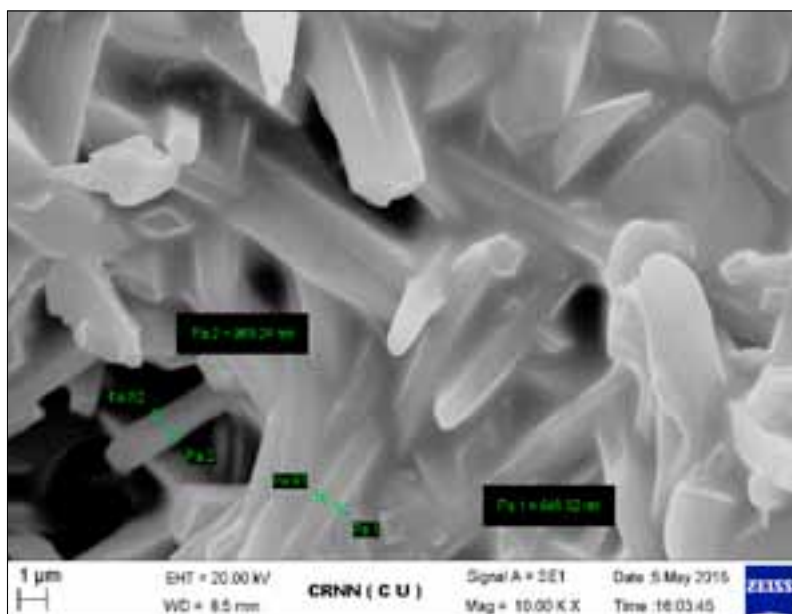
as major crystalline phase along with cristobalite (PDF No 82-1409) as minor phase. Cristobalite phase is found to be gradually disappeared while mullite crystallinity increases with progressive rise in temperature up to 1575 °C as indicated by sharp rise of relative intensity of characteristic peaks. This may be explained by dissolution of in situ alumina with transitory liquid phase leading to the precipitation of secondary mullite crystals.

In presence of synthetic boehmite and 1400 °C onwards, rapid mullitisation occurs by dissolving the reactive alumina and cristobalite in to glassy phase leading to secondary mullite formation by solution precipitation mechanism. It is presumed

that in presence of reactive alumina, random mullite nucleation tends to occur. The XRD patterns (Fig. 6) of boehmite added bodies confirm that with increasing addition of boehmite, more and more mullitisation occurs resulting in enhancement of densification. With increase of temperature of heat treatment, mullite phase becomes more prominent with less or no cristobalite content.

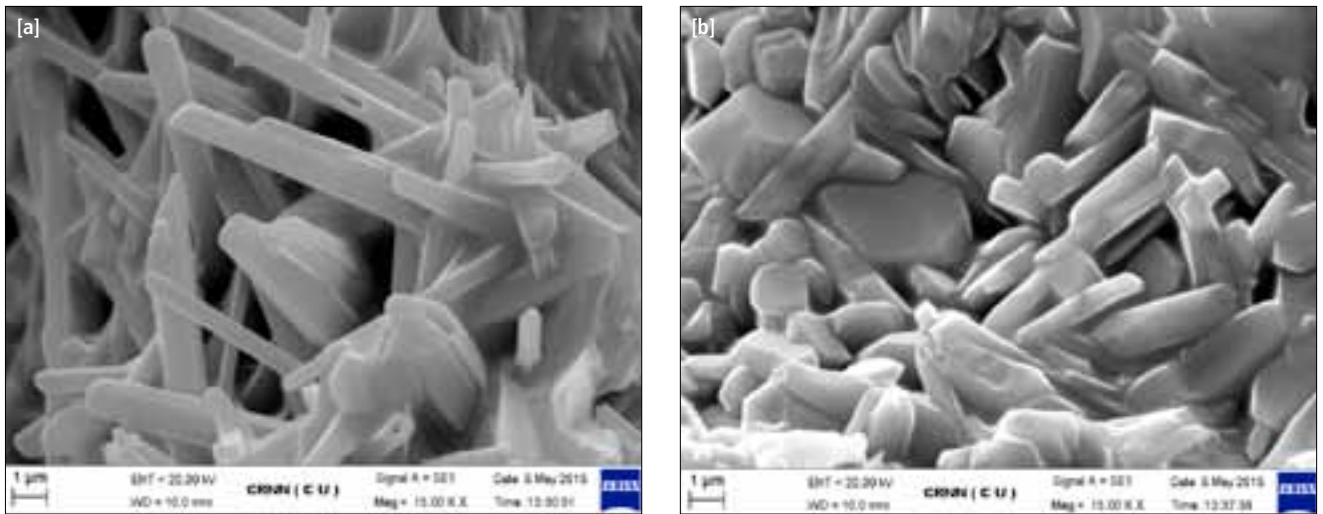
The microstructure of the etched surface of the samples sintered at different temperatures was recorded by scanning electron microscope. Few cuboidal shaped primary mullites (MI) are observed in the glassy matrix generated from interaction of impurities, fly ash and precipitated silica in batch-1 bodies sintered at 1400 °C. Fig. 7 a also reveals that the structure is highly porous in nature contributing low bulk density. In the present study, glass phase appears comparatively at lower temperature probably due to the dissolution of transition metallic oxides like  $\text{Fe}_2\text{O}_3$ ,  $\text{TiO}_2$  or alkali, alkaline-earth oxides (MgO, CaO, etc.) associated with the starting materials.

When sintered at 1450 °C (Fig. 7 b), grain morphology appears to be bimodal in nature consisting of equiaxed secondary mullite with smaller aspect ratio and cuboidal primary mullite with rounded corners. It is proved that the  $\text{Al}_2\text{O}_3$ – $\text{SiO}_2$  ratio of cuboidal primary mullite is close to 2 : 1. The rounded corners of the grains depict uneven growth of mullite from liquid phase. The secondary mullite is thought to be nucleated from the transitory glassy phase formed by dissolution of in situ alumina from bauxite. The presence



**Fig. 8** SEM micrographs of batch-1 samples sintered at 1575 °C





**Fig. 9 a–b** SEM micrographs of samples sintered at 1450 °C: [a] batch 2B, [b] batch 2D

of less glassy phase with compact microstructure yields higher bulk density value. With enhancing the sintering temperature from 1450 °C to 1575 °C, more elongated mullite grains with well interlocked structure and longitudinal growth is observed (Fig. 8). The SEM reveals a microstructure with uniform distribution of grains in the nano size range from 850–970 nm.

The aspect ratio in secondary mullite is mainly controlled by the amount of liquid formed at sintering temperatures, viscosity of liquid and the duration of the crystallization taking place. Acicular shape (MIII type) mullite has been formed due to the anisotropic solid-to-liquid surface energy between the mullite crystal and the eutectic liquid [29].

The acicular shape of crystal is precipitated from the matrix due to presence of dopant, which lowers the surface energy of the crystal in certain directions, leading to a preferential growth. Addition of boehmite generates elongated columnar-acicular mullite crystals with varying width of 600–950 nm (Fig. 9 a).

With higher percentage incorporation of boehmite, enhanced formation of MII-type secondary mullite (Fig. 9 b) with lower aspect ratio along with few MIII-type secondary mullite are found. Microfine active  $Al_2O_3$  from boehmite augments the solid state interdiffusion rate [30] and thereby facilitating the growth of the mullite seeds. The boehmite addition to the mullite batches favours uniform size distribution of mullite grains which result in maximization of density and reduction of porosity [31] without

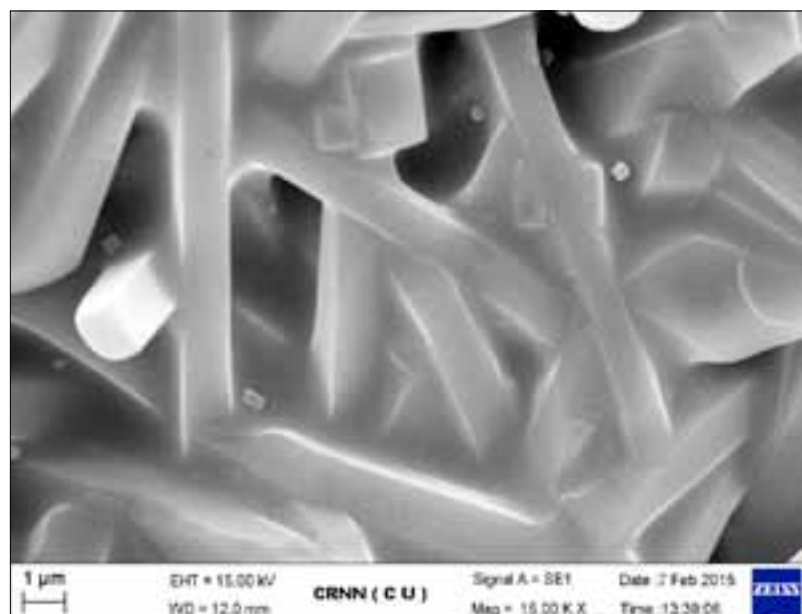
much grain growth. Fig. 10 showed existence of corundum grains (~40 nm) adhered to contact points of mullites favouring diffusion of ions with growth in the neck areas thereby filling the pores.

**Conclusion**

From this investigation it can be concluded that

- Cost-effective mullite ceramics is evolved at a relatively lower sintering temperature using indigenous starting materials: natural bauxite, fly ash and precipitated silica. The effect of doping action of synthetic boehmite on the densification of mullite is well evidenced.

- Without addition of dopant, primary mullite along with cristobalite are preferentially formed as crystalline phases during lower temperature of sintering at 1400 °C which on rise of sintering temperature up to 1550 °C, augments the formation of well-crystalline secondary mullite with the progressive elimination of quartz and primary mullite in the sintered body.
- The significant improvement in thermo-mechanical properties and dense microstructure in in-situ formed mullite bodies containing 1,2 % boehmite is achieved during sintering at around 1550 °C.
- During firing around 1550 °C, minor presence of boehmite accelerates precipita-



**Fig. 10** SEM micrograph of batch-2D sample sintered at 1575 °C

tion mechanism leading to the formation, growth and uniform distribution of anisotropic acicular secondary mullite grains in the well-sintered compacts.

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