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Research paper

Effect of temperature on mullite synthesis from attrition-milled pyrophyllite and α-alumina by spark plasma sintering

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ABSTRACT

This paper addresses the effects of processing technique on the synthesis of mullite from pyrophyllite and alpha-alumina powder. The influence of firing temperatures were examined in order to optimize the fabrication process to achieved a suitable mechanical properties. Feedstock powders, with stoichiometric composition, were wet milled in an Attritor mill. The dried powders were consolidated using spark plasma sintering (SPS) in the temperatures range 1400 °C to 1700 °C under pressure of 50 MPa, with a heating rate of 100 °C/min at different holding times. Densities of 3.25 g/cm^3 and 3.17 g/cm^3 were obtained for the samples sintered at 1400 °C and 1600 °C respectively, with a 10-min holding time. The XRD and SEM/EDS were employed to characterize the firing transformation and microstructure of the samples. The SEM micrograph of samples, sintered at 1600 °C with 10 min holding times, revealed that the sample retained its equiaxed grain structure. The XRD results show a reduction in alpha-alumina content after the sample was fired. Hardness and fracture toughness values up to 11.73 GPa and 1.99 MPa•m$^{1/2}$ respectively were obtained.

1. Introduction

Mullite (Al_6 Si_2O_{13}) has emerged as the material of choice for refractories and high- technology applications in optical, electronic and structural components (Aguilar Santillan et al., 2007). This is due to its high melting point, very good creep resistance, high shear modulus and good corrosion resistance (Kanka and Schneider, 1994; Schneider et al., 2008). However, mullite rarely occurs in nature due to its high temperature and pressure formation conditions (Raghdi et al., 2017). In the field of material science, a significant advantage is gained when different materials are combined to achieve a new set of desired properties. In order to convert pyrophyllite to mullite, alumina was added for the preparation of the specimens, with the 3:2 Al_2O_3: SiO_2 mullite stoichiometry.

Previous studies have shown that the formation of the stoichiometric composition and the phase of mullite depend on the synthesis technique, as well as the kind of raw material employed (Pani et al., 2015). Several research reports have been published on the synthesis of mullite using different approaches such as sol-gel (Amutharani and Gnanam, 1999), co-precipitation (Chaudhuri and Patra, 1997), hydrolysis (Pask et al., 1987) and reactive sintering (Aksay and Pask, 1975). So, Yu et al. (2014), reported on the thermal reaction of cristobalite in a nano-SiO_2/αAl_2O_3 powder system for mullite synthesis. Similarly, Lee et al. (2002) investigated the effect of precursor pH and the sintering temperature on the synthesized sol-gel processed mullite. Although these processes yield high purity mullite, the disadvantages of these approaches are the long processing time and high cost of starting materials which are not appropriate for large scale production (Pani et al., 2015; Viswabaskaran et al., 2002). Pani et al. (2015) reported on the formation of mullite from an aluminous-rich mine waste (shale) from Fe/Mn mines using thermal plasma reactor for only 5 min. In addition, Kanka and Schneider (1994), reported on the sintering mechanism and microstructural development of co-precipitated mullite. Homogenous and dense materials consisting of equiaxed mullite grains in their microstructure together with a small amount of α-Al_2O_3 were produced from high-Al_2O_3 powders calcined at 600 °C and 1100 °C. Mullite derived from natural minerals such as Kaolinite, kyanite, sillimanite, andalusite and aluminous-rich rock or clay, with the addition of alumina, has been studied for different purposes (Aguilar Santillan et al., 2007; Chen et al., 2000; Viswabaskaran et al., 2002; Pani et al., 2015). Wang et al. (2010), had also reported on the synthesis of mullite from waste aluminium slag and pyrophyllite fabricated by conventional sintering route.

However, there is virtually no reported study available on Spark Plasma Sintering (SPS) of pyrophyllite and reactive alumina to fabricate mullite and their post heat treatment behavior. The Spark Plasma

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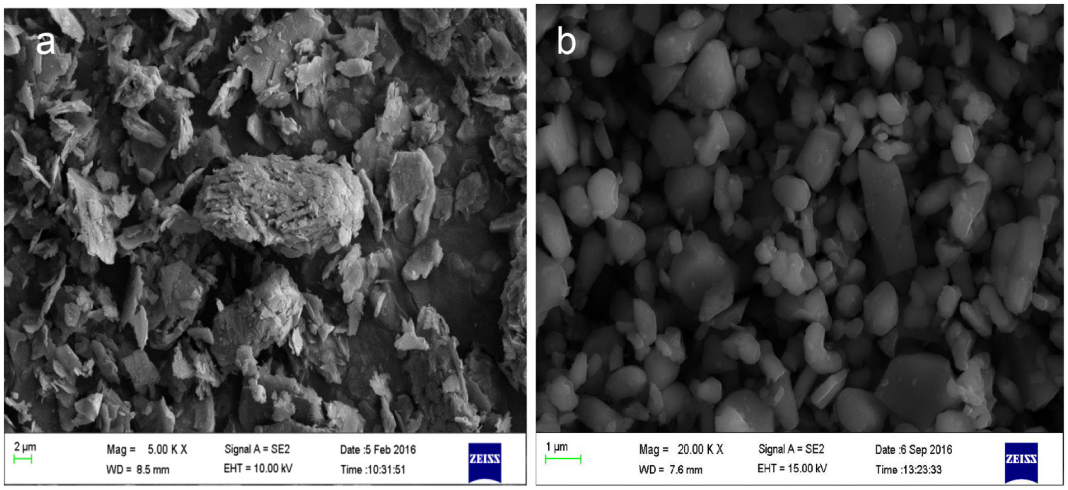

Fig.1. (a–b): SEM micrograph of as-received powders (a) pyrophyllite (b) alumina.

Table 1
Chemical composition (wt%) of the as received pyrophyllite powder.

Mineral	SiO_2	Al_2O_3	Fe_2O_3	FeO	MnO	MgO	CaO	Na_2O
wt%	58.63	30.30	0.12	0.99	0.01	0.07	0.05	0.17
Mineral	K_2O	TiO_2	P_2O_5	Cr_2O_3	NiO	LOI	Total	
wt%	1.09	1.94	0.15	0.03	0.00	6.06	99.60	

Sintering (SPS) process has been used in the fabrication of transparent mullite (Zhang et al., 2009). The accurate control of sintering conditions as well as high sintering speed makes SPS a promising technique for producing high dense materials with controlled grain growth (Suárez et al., 2013). An effort is therefore made in this study to develop mullite from locally mined pyrophyllite with addition of α-alumina powder using SPS. The sample density, microstructure, hardness and fracture toughness were investigated.

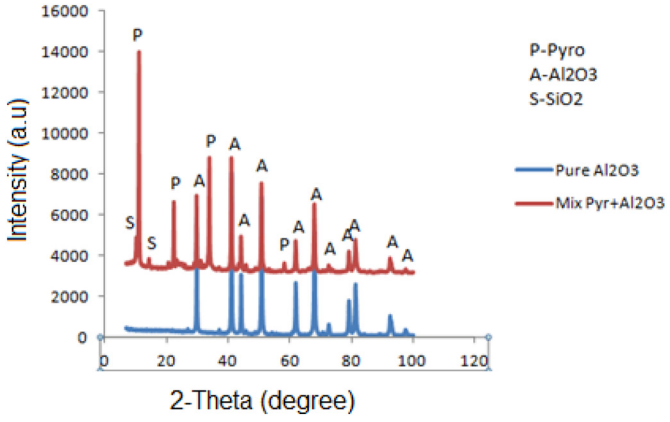

Fig. 3. XRD stack of pure alumina and mixture of pyrophyllite and alumina powder.

2. Materials and methods

2.1. Sample preparation

Pyrophyllite powder, mined in Ottosdal in South Africa, and reactive α-alumina (P172SB) of 99.7% purity (CERadvance Engineering Ceramics (Pty) Ltd., South Africa), with a particle size of 0.4 µm were combined to prepare mullite specimens. The as-received pyrophyllite powder has a particle size of < 63 µm. Scanning Electron Microscopy (SEM) and powder X-Ray Diffraction (RXD) were used to characterize the morphology of the particles and identify the phases present in the as-received powders. PANalytical X-ray fluorescence (model AXIOS mAX spectrometer at 50 kV and 50 mA) was used to characterize the chemical composition of as-received pyrophyllite powder. The removal of any organic residues was done by placing the Pt crucibles and LOl dish in a furnace at 1020 °C for half an hour. Approximately 1 g of powder sample was placed in LOI dish and Ignited at 1020 °C for 40 min. The analysis was performed by superQ software. The as-received pyrophyllite powder was then milled down to 0.5 µm and fired in a tube furnace at 800 °C for 30 min to produce a dehydroxylated pyrophyllite powder. Stuckey (1924), studied the dehydroxylation of pyrophyllite as a function of temperature and found that when heat-treating this material at 800 °C for 15 min, pyrophyllite was largely dehydrated and the resulting material contained 0.2 wt% of water which was driven off at 900 °C. The ratio of fired pyrophyllite and alumina powder in stoichiometric mullite composition formulation was expressed as:

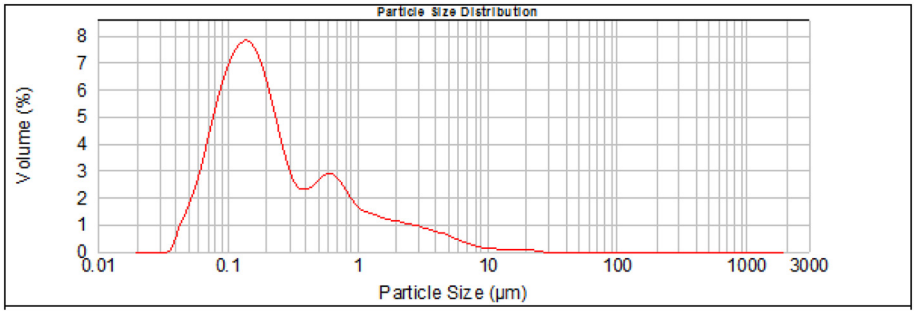

Fig. 2. The Particle size distribution of alumina and pyrophyllite powder milled for 6 h.