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Ceramic compositions based on nano forsterite/nano magnesium aluminate spinel powders

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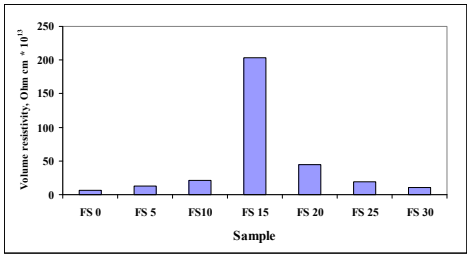
HIGHLIGHTS

- Ceramic compositions based on nano forsterite/nano-$MgAl_2O_4$ spinel were synthesized.
- CCS was improved (333.78 MPa) through 15 mass% of nano-$MgAl_2O_4$ spinel addition.
- Volume resistivity was enhanced to $203*10^{13}$ Ohm cm with 15 mass% of spinel addition.
- Beyond 15 mass% spinel, CCS and volume resistivity were decreased.

GRAPHICAL ABSTRACT



Volume resistivity of the samples FS0-FS30 fired at 1550°C.

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ABSTRACT

According to the wide applications in the field of chemical and engineering industries, forsterite (Mg_2SiO_4)/spinel ($MgAl_2O_4$) ceramic compositions were the matter of interest of several research works during the last three decades. This work aims at preparation and characterization of improved ceramic bodies based on forsterite and spinel nano powders through controlling the forsterite and spinel contents in the prepared mixes. These prepared ceramic compositions have been investigated through measuring the densification parameters, cold crushing strength as well as volume resistively. Nano spinel was added from 0 to 30 mass% on expense of nano forsterite matrix and fired at 1550 °C for 2 h. The phase composition of the fired samples was examined using x-ray diffraction (XRD) technique. The microstructure of some selected samples was shown using scanning electron microscope (SEM). A pronounced improvement in the sintering, mechanical properties and volume resistively were achieved with increasing of nano spinel addition up to 15 mass%. This is due to the improvement in the matrix of the prepared forsterite/spinel bodies as a result of well distribution of spinel in the forsterite matrix as depicted by SEM analysis.

1. Introduction

Nanoparticles offer a host of attractive properties which includes increased strength, high hardness, high diffusion rates, and reduced sintering times in comparison to those associated with coarser particles [1–3]. Forsterite is a crystalline magnesium silicate with chemical formula Mg_2SiO_4, named after the German naturalist Johann Forster. It belongs to the group of olivine. Like clinoenstatite, with the formula $MgSiO_3$, forsterite has an extremely low electrical conductivity [4]. This makes forsterite ceramics an ideal substrate material for electronics. It is a material of interest to

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engineers and designers, especially as an active medium for tunable laser [5–7], owing to its properties like good refractoriness, with the melting point at 1890 °C, low dielectric permittivity, thermal expansion, chemical stability as well as excellent insulation properties even at high temperatures [8]. Sano et al. [9] reported some fabrication techniques that allow high density aggregate of forsterite. Also, they reported its dielectric properties [9].

Various composites were developed to improve the physicomechanical properties, strength, dielectric constant, thermal expansion, thermal conductivity as well as mechanical performance of forsterite ceramics. The properties are improved through addition of magnesium aluminate spinel, alumina, mullite, and partially stabilized zirconia [10]. Magnesium aluminate spinel ($MgAl_2O_4$) is one of the best known and widely used polycrystalline materials on account of its attractive properties such as high melting point, high mechanical strength at elevated temperatures, high chemical inertness, and good thermal shock resistance [11–13]. These special properties make $MgAl_2O_4$ a potential candidate as a refractory material [11–13] and catalyst support [14,15]. Recently, others different studies on the catalyst support have been developed [16–18]. Mustafa et al. [10] investigated the effects of spinel phase on the mechanical properties of forsterite ceramic. They observed that the formation of spinel phase strongly improved cold crushing strength of forsterite bodies. However, Koizumi et al. [19] showed the sintering result of forsterite phase plus spinel phase.

Forsterite-spinel composite has been synthesized by different methods such as solid–state reaction, self-propagation high temperature synthesis, and sol–gel. The production of forsterite-spinel composite via solid–state reactions usually requires high temperature and long reaction time. This is mainly because the reaction of the starting oxides is generally slow due to the relatively low diffusivity which results in the formation of enstatite ($MgSiO_3$) and/or MgO instead of forsterite. Hence very high processing temperature of 1200–1600 °C is required [20] resulting in coarse grained powders while the sol–gel process can provide molecular-level of mixing and high degree of homogeneity, which leads to reduce crystallization temperature and prevent phase segregation during heating [21–24].

Little works have started with nanopowder for prepare forsterite/spinel ceramic compositions, so this work aims at preparation and investigation of improved ceramic composites based on nano forsterite and nano spinel powders.

2. Materials and experimental

The starting materials used in this study were magnesium chloride hexahydrate ($MgCl_2.6H_2O$, Mw = 203.30 g/mol, S.D. Fine – Chem. Ltd., India) and tetraethyl orthosilicate (TEOS, $C_8H_{20}O_4Si$, Mw = 208.33 g/mol, Merck Schuchardt OHG, Hohenbrunn, Germany) for nano forsterite synthesis and aluminum chloride hexahydrate ($AlCl_3.6H_2O$, Mw = 241.43 g/mol, Merck Schuchardt OHG, Hohenbrunn, Germany) and magnesium chloride hexahydrate for spinel synthesis.

Nano forsterite and nano spinel powders were synthesized via a modified sol–gel and co-precipitation techniques, respectively with average crystalline size 20–33 nm and average particle size ~70 nm, as noted by the authors [25].

Seven batches were prepared from different mixes of nano forsterite with different additions (0–30 mass%) of nano spinel powders (designated as FS0, FS5, FS10, FS15, FS20, FS25 and FS30), Table 1, and pressed using uniaxial pressing machine at 125 MPa. The obtained pellets were then sintered at 1550 °C, for 2 h in air.

Phase analysis was shown by X-ray diffractometry (XRD) (Philips 1730 diffractometer with a Ni-filtered Cu-Kα radiation at a scanning speed of 1°/min).The sintering parameters in terms of bulk density and apparent porosity of the fired compacts were determined according to ASTM-C20 method. This method based on drying the test samples to constant weight by heating to 110 °C overnight and determine the dry weight, Wd, in grams. Then, place the test samples in water and boil for 2 h. During the boiling period, keep them entirely covered with water. After the boiling period, cool the test specimens to room temperature while still completely covered with water. Determine the suspended weight, Wi, of each test sample after boiling and while suspended in water in grams. After determining the suspended weight, blot each sample lightly with a moistened smooth cotton cloth to remove all drops of water from the surface and determine the saturated weight, Ws, in grams. The bulk density, BD, and the apparent porosity, AP, of the test samples were calculated according to the following equations:

Table 1
Batch design of the investigated samples FS0–FS30/(mass, %).

Sample	Nano forsterite	Nano spinel
FS0	100	0
FS5	95	5
FS10	90	10
FS15	85	15
FS20	80	20
FS25	75	25
FS30	70	30

Apparent Porosity, AP

$$AP = \frac{\mathbf{Ws} - \mathbf{Wd}}{\mathbf{Ws} - \mathbf{Wi}} \times 100$$

Bulk density, BD

$$BD = \frac{\mathbf{Wd}}{\mathbf{Ws} - \mathbf{Wi}} \times \gamma$$

Wd: weight of dry sample in air.
Wi: weight of suspended sample in water.
Ws: weight of saturated sample in air.
γ: specific gravity of water

The total linear shrinkage of the bodies was determined by measuring the length of the samples before and after the sintering process. Cold crushing strength (CCS) of the fired bodies was determined using a hydraulic testing machine according to ASTM: C133-97. Each of these tests was carried on three samples of each mix taking the average values. The microstructure of some selected fired samples was examined using scanning electron microscope (Philips XL 30) of gold coated fractured samples. The volume resistivity of the sintered samples was measured by four-probe method using Keithley 6517B electrometer.

3. Result and discussion

3.1. Densification of forsterite/spinel compositions

The bulk density (BD) and apparent porosity (AP) of the sintered ceramic bodies FS0–FS30 fired at 1550 °C are shown in Fig. 1. A noticeable improvement in the densification parameters of the sintered samples containing 15 mass % of nano-spinel addition in comparison with others samples have been observed. This is generally due to: 1) the effect of nanoparticles on the sintering of ceramic bodies which yields a high sintering and excellent grain