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Microwave synthesis and sintering of forsterite nanopowder produced by high energy ball milling

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Abstract

This paper reports the development of a new process for the synthesis and sintering of forsterite nanopowder via microwave-assisted high energy ball milling of a powder mixture containing silica gel and $Mg(OH)_2$. X-ray diffraction (XRD), FTIR spectrometer, BET, scanning electron microscopy (SEM) and Transmission electron microscopy (TEM) techniques were utilized to characterize the as-milled and annealed samples. X-ray diffraction results showed that highly ordered forsterite can be obtained through the calcination of the as-milled powder over 900 °C. In addition, SEM and TEM observations of the synthesized powders showed that the particle size of the powder lies in the nanometer range, also being compared with the BET results (about 45 to 64.5 nm). Microwave sintering (MS) of the forsterite nanopowder produced with high energy ball milling and subsequent microwave heating resulted in remarkable enhancement in densification in comparison with conventional sintering (CS) at lower temperatures.

Keywords: A. Sintering; Microwave heating; Synthesis; Forsterite; High energy ball milling

1. Introduction

Modern technologies constantly require materials with special properties to achieve breathtaking innovations. This, in turn, requires constant improving of scientific and technological fabrication as well as working procedures. Nanocrystalline materials are single-phase or multi-phase materials, whose crystal size is in the order of a few (typically 1–100) nanometers at least in one dimension. Because of the extremely small size of the grains, a large fraction of the atoms in these materials is located in the grain boundaries and thus the material exhibits enhanced combinations of physical, mechanical, and magnetic properties (compared to material with a more conventional grain size, i.e., >1 μm). Therefore, nanocrystalline materials show increased strength, high hardness, extremely high diffusion rates, and consequently reduced sintering times for powder compaction [1]. Forsterite is a crystalline magnesium silicate with chemical formula Mg_2SiO_4, named after the German naturalist Johann Forster. Forsterite (Mg_2SiO_4) is an important material of olivine family of crystals in the magnesia–silica system with orthorhombic structure [2]. The extremely low electrical conductivity of forsterite makes it an ideal material for tunable laser [3–5]. Moreover, it shows good refractoriness with high melting point (1890 °C), low thermal expansion, good chemical stability and excellent insulation properties even at high temperatures [6–8]. The manufacturers of the SOFC (solid oxide fuel cells) find forsterite interesting due to its linear thermal expansion coefficient perfectly matching the other cell components and a very high stability in fuel cell environments [9]. In the recent years, much attention has been paid to the development of microwave telecommunication technologies because of the increased requirements for microwave applications. Among these materials, forsterite Mg_2SiO_4 has attracted a great deal of attention due to its low dielectric constant and loss tangent [10,11]. Furthermore, forsterite bioceramic possesses good biocompatibility and mechanical properties and might be suitable for hard tissue repair [12–14]. Various techniques including the heating of mixed powders prepared by the alkoxy method [15], the polymer matrix method [16], the citrate–nitrate route [17], the sol–gel method [18–20], the

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combustion method [21] and high energy ball milling have been used to synthesize forsterite [22–24]. It is been recently found that microwave energy can be used to synthesize ceramic powders where reactions of component oxides at elevated temperatures are involved [25]. Microwave synthesis of materials is fundamentally different from the conventional synthesis in terms of its heating mechanism. In the microwave furnace, the interaction of microwaves with the material generates heat within the sample volume [26]. Moreover, microwave energy heats the material on a molecular level which leads to uniform heating, whereas conventional heating systems would heat the sample up from the surface towards the interior zones, giving rise to steep thermal gradients [27]. The microwave assisted preparation of nanopowders is a new method that includes the hydrothermal, hydrolysis and co-precipitation methods [28–30]. In this paper, a novel approach which is the combination of ball milling and microwave heating for the synthesis and sintering of forsterite has been developed.

2. Experimental

2.1. Materials

Silica gel and magnesium hydroxide ($Mg(OH)_2$) were used as the starting materials for the synthesis of forsterite. Figs. 1 and 2(a, b) show the XRD and morphology of the initial powder agglomerates. The XRD pattern of raw materials was characterized according to those of silica gel (amorphous pattern) and $Mg(OH)_2$ (XRD JCPDS data reference code 007-0239). The $Mg(OH)_2$ powder has an angular shape with a mean agglomerate diameter of about 5 μm. Silica gel powder has a spherical shape with a mean agglomerate size of about 1–2 μm. The agglomerate size of powders was determined using scanning electron microscope (SEM). Also several micrographs were used for agglomerate size measurement and the average value was reported.

2.2. Experimental procedure

The milling experiment was carried out with Planetary Mill. A zirconia vial with diameter of 80 mm and 25 zirconia balls with diameter of 15 mm were used as the milling medium. The required amount of powder mixture for 15:1 ball to powder mass ratio (BPMR) was taken from the homogeneous mixture of powders and placed in the bowl for ball milling. The raw materials were milled in air at room temperature for 0.25, 5, 10, 20, 30 and 40 h, respectively. The rotation speed of the disk was 270 rpm and that of the vials was 675 rpm. The milled powders were calcined at 500–1200 °C by microwave heating in the air. Powders pre-pressed up to ~49.5% of the theoretical density were sintered in two different ways: (i) conventional sintering (CS) via continuous heating up to different temperature (1150–1350 °C) with a heating rate of 10 °C/min, (ii) microwave sintering (MS) through heating up to the same temperature with conventional sintering in a 1.1 kW, 2.45 GHz multimode microwave cavity (Bosch, Germany).

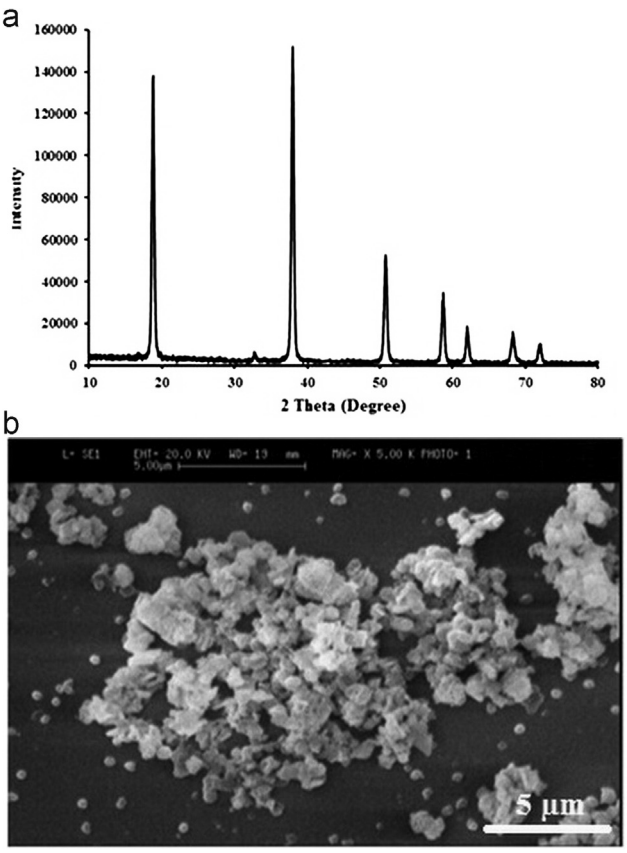

Fig. 1. (a) XRD and (b) SEM $Mg(OH)_2$ powder before high energy ball milling.

2.3. Characterization

The structural properties of the samples were investigated by X-ray diffraction (Simens D-500 system) technique using a CuKα monochromatized radiation source and Ni filter in the range 2θ=10–80. The morphology, microstructure and the particle size of the high energy ball milled powders and sintered samples were examined by a Philips scanning electron microscope (SEM) operating at 20 kV and Transmission electron microscopy (TEM, TecnaiF20, Phillip, Holland). The density of the sintered samples was measured according to the Archimedes method. FTIR spectroscopy of the test materials was carried out by a Fourier transform infrared spectrometer (Bruker, V33 spectrophotometer) from 400 to 4000 cm^{-1}, using KBr pellets containing 1% weight sample in KBr. Also, the surface area (BET) was determined by nitrogen adsorption at −196 °C using an automated gas adsorption analyzer (Micrometrics, Gemini 2375). The BET surface area was used to calculate the mean particle size [D] (Eq. (1)).

$$D = 6/s\rho \quad (1)$$

where 's' is the BET surface area (m^2/g) and 'ρ' is the density of forsterite (kg/m^3). The density of forsterite was considered as 3.27×10^3 (kg/m^3).