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Production of dense forsterite with ultra low dielectric loss using ZrO_2 additive

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ABSTRACT

Forsterite was synthesized with $MgCO_3$ and SiO_2 powders using a solid-state method at 1200–1300°C. The ceramic specimen with the highest forsterite phase content was sintered with the ZrO_2 sintering aid at mass fractions of 0.5, 1.0, and 1.5 wt% at 1400°C for 4 h. The dielectric constant and loss tangent were also measured via the waveguide method at 8–12 GHz. The formation of enstatite and cristobalite was minimized through precise synthesis temperature control. Phase measurement by the Rietveld method showed that specimen F28 contained more than 99.8% forsterite. It was found that ZrO_2 reduced the pore volume percent and increased the density even at low mass fractions. A liquid phase on the grain boundaries was demonstrated. The solid ceramic specimen F28-15Z showed a dielectric constant of 6.5 and high quality factor of 189000.

1. Introduction

Forsterite (chemical formula: Mg_2SiO_4) is a magnesium-rich mineral composed of SiO_4^{-4} anions and Mg^{2+} cations in a molar ratio of 1:2. It has a high melting point (1890°C), high creep resistance, low thermal conductivity (2.8×10^{-5}-4.5×10^{-5} C^{-1} from 27°C to 1870°C), low dielectric permittivity (67–72 at 1 MHz), and high chemical stability [1–3]. Pure forsterite has 45% ionic bonding and 55% covalent bonding. The high strength of covalent bonds reduces the dielectric constant so that it approximately equals $\varepsilon_r = 6.5$ for forsterite [4,5]. Silicate compounds with dielectric constants below 10 are good candidates for directing millimeter waves. These waves have a high data transmission rate and are applied for radar systems. Wireless communication systems require materials with a high-quality factor and a low-loss tangent (tanδ = 1/Q) [6]. Pure forsterite with a high-quality factor (Q.f) is an important compound for infrared (IR) and millimeter wave applications [7]. Ohsato et al. [8] showed that forsterite is among the ideal materials for this purpose. Forsterite can also be employed in high-frequency insulators [9], tunable lasers [2], and solid oxide fuel cell (SOFC) interlayers [3].

There has been a great interest in the synthesis of pure forsterite. Several methods have been developed to synthesize forsterite, including solid-state synthesis, sol-gel, mechanical activation, polymer synthesis, and co-precipitation [10]. Among them, the direct reaction between MgO and SiO_2 at a temperature up to 1525°C is the most common method for forsterite synthesis because it is more facile and preferable for large scale production [11,12]. The use of precursors such as $Mg(OH)_2$ and $MgCO_3$ in forsterite synthesis results in an additional intermediate reaction. The precursors should first decompose before Mg reacts with SiO_2. Cheng et al. [13] employed serpentine that contains MgO and SiO_2 to synthesize forsterite. The precursors were mixed in the stoichiometric ratio of forsterite and subjected to mechanical alloying. Despite this additional reaction (decomposition of magnesium carbonate), the results showed that $MgCO_3$ and $Mg(OH)_2$ had higher reactivity than MgO, reducing the formation temperature of forsterite [14].

However, solid-state synthesis has some disadvantages, including the low density of the synthesized product due to the formation of unwanted phases such as $MgSiO_3$ and residual MgO [15]. Furthermore, some studies have reported the formation of $Mg_2SiO_4/MgSiO_3$ layers between Mg and SiO_2 particles [16].

Many techniques have been developed to accelerate and complete the formation of forsterite, *e.g.*, activating precursor powders through mechanical alloying. Mechanical alloying improves the mixing of precursors and accelerates the synthesis reactions during heat treatment and sintering [17].

On the other hand, researchers have sintered forsterite using different methods to fabricate monolithic products after synthesizing forsterite. Pressureless sintering, spark plasma sintering (SPS), and hot

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