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Synthesis and microwave dielectric properties of $Li_2Mg_2(WO_4)_3$ ceramics

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ABSTRACT

Lyonsite-structured $Li_2Mg_2(WO_4)_3$ (LMW) ceramics with low loss were prepared via a conventional solid-state method. Synthesis process, microstructure and microwave dielectric properties of LMW were systematically investigated. The results indicated that higher calcination temperature and addition of excess Li effectively eliminated the secondary phase $MgWO_4$, and pure orthorhombic LMW phase could be prepared at 900 °C for 4 h using an excess amount of 1 mol% Li_2CO_3. Optimum microwave dielectric properties of $\varepsilon_r=8.2$, $Q\times f=90{,}000$ GHz ($f=11.2$ GHz), and $\tau_f=-52.4$ ppm/°C were obtained when pure LMW was sintered at 800 °C for 2 h. Moreover, for the $(1-x)$ LMW–xTiO$_2$ samples, superior microwave properties of $\varepsilon_r=12.2$, $Q\times f=50{,}000$ GHz ($f=9.6$ GHz) and especially a near-zero τ_f (-2.8 ppm/°C) could be achieved when the $x=0.14$ (in volume ratio) sample was sintered at 850 °C for 2 h.

1. Introduction

Dielectric oxide ceramics have revolutionized the microwave wireless communication industry [1]. Up to date, numerous dielectric ceramics with suitable relative permittivity (ε_r), high quality factor ($Q\times f$), and near-zero temperature coefficient of resonant frequency (τ_f) have been reported. However, most of them have relatively high sintering temperatures, which significantly restrict their further applications. To reduce the sintering temperature of dielectric ceramics, materials with inherently low sintering temperature have attracted considerable attention, such as V_2O_5-, MoO_3-, WO_3- based systems [2–4].

Lyonsite [5] is a ubiquitous type of adaptive structure. Zhou et al. [3] have reported a lyonsite family of molybdates, which show good microwave dielectric properties with relatively low sintering temperature. In fact, the lyonsite-type compounds also exist in tungstates and vanadates. As far as we know, there have been only a few researches involved in such materials to date. In the Li_2O–MgO–WO_3 system, the lyonsite structure of $Li_2Mg_2(WO_4)_3$ (LMW) was first reported by Fu et al. [6] Recently, Guo et al. [7] reported that stoichiometric LMW exhibited good microwave dielectric properties ($\varepsilon_r=7.72$, $Q\times f=29{,}600$ GHz, and $\tau_f=-15.5$ ppm/°C) when it was synthesized at 750 °C for 3 h and sintered at 875 °C for 3 h. It is also chemically compatible when cofired with Ag. Surprisingly, a three times higher $Q\times f$ value of ~90,000 GHz could be achieved in current work when a similar solid-state route was used. To explore the potential causes of the significant improvement of $Q\times f$, the synthesis, sintering behavior, microstructure and microwave dielectric properties of LMW were systematically investigated. In addition, a near-zero τ_f value was further obtained by adding a few amount of TiO_2.

2. Experimental

Ceramics were prepared by a conventional mixed oxide route using the high-purity (>99%) powders of MgO, WO_3 and Li_2CO_3. Stoichiometric LMW powders with and without excess 1 mol % Li_2CO_3 were weighed, and ball-milled in alcohol medium for 4 h. The wet mixture was rapidly dried and then calcined at 750 °C, 800 °C and 900 °C for 4 h. Afterwards, pure LMW powders, obtained with excess lithium calcined at 900 °C, were ball-milled for 6 h. The granulated powders were mixed with 5 wt% polyvinyl alcohol (PVA) as a binder and subsequently pressed into cylinders with dimensions of 10 mm in diameter and 5–6 mm in height, and finally sintered at 700–900 °C for 2 h. To tailor its large negative τ_f, 14–16 vol% TiO_2 were mixed with pre-synthesized pure-phase LMW powders, and then sintered at 85 °C for 2 h.

The crystalline phases were identified by an X-ray diffractometer (XRD; D/Max2500V, Rigaku, Tokyo, Japan). A least mean square method of analysis was used to calculate lattice parameters with Jade 6.0 software. The bulk densities of the sintered ceramics were measured using the Archimedes method. The

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