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Synthesis and microwave dielectric studies of pure Li_2MgSiO_4 and B_2O_3, MgF_2, WO_3 added Li_2MgSiO_4 for substrate applications

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ABSTRACT

Synthesis of pure Li_2MgSiO_4 (LMS) ceramics through solid-state ceramic route and variations in its microwave dielectric properties by addition of different compounds are discussed. Boron trioxide (B_2O_3), Magnesium Fluoride (MgF_2), and Tungsten trioxide (WO_3) were used as additives to produce three different ceramics besides pure LMS. Scanning electron microscope and X-ray diffraction spectrometer were used to study the morphology and crystal structure of prepared samples. The densities of the respective samples were measured via Archimedes method. The microwave dielectric properties of the ceramic materials were determined by the cavity perturbation technique using a Scalar Network Analyzer. Pure LMS sintered at 1100 °C/6 h exhibited a relative permittivity (ε_r) of 5.73 and dielectric loss of 5.897×10^{-4} at 8 GHz. Among the three additives, WO_3 improved the relative density of pure LMS from 90.6% to 94.5% and the LMS-WO_3 sample sintered at 1100 °C/4 h showed comparatively good dielectric properties, $\varepsilon_r = 6.03$, $\tan\delta = 6.598 \times 10^{-4}$ at 8 GHz.

1. Introduction

Significance of microwave in the communication domain has been increasing after its application in radar during World War II. Transmission of energy and information signals without interference which is barely possible by radio waves, line of sight propagation, wavelength similar to the circuit dimensions, higher frequencies allowing more storage than radio waves and smaller antenna sizes, short wavelength producing large reflections, ability to be focused into narrow beams, etc multiplied the use of microwaves in point-to-point telecommunications, satellite communication, television, radar, military telemetry, remote sensing, tracking applications etc. In the electromagnetic spectrum the microwaves reside in between radio and infrared waves with a frequency span of 300 MHz–300 GHz and a wavelength range of 0.1 cm–100 cm. Even though different sources assign different frequency range for microwaves, it always includes the whole Super High Frequency (SHF) band (3–30 GHz, 10–1 cm). According to the IEEE standard S, C, X, K_u, K, and K_a are referred as the microwave frequency bands and these are mainly utilized for satellite communication. Expansion of microwave telecommunication and satellite broadcasting industry led to the development of a variety of microwave devices which have become obligatory in microwave communication systems. Therefore the microwave dielectric ceramics have been comprehensively explored for microwave device applications, such as resonators, filters, and oscillators [1,2]. Their broad range of applications includes cell phones, GPS, antennas, packages, substrates, capacitors, filters, resonators etc [3–6]. The three key parameters that should be considered for the substrate applications are relative permittivity, dielectric loss, and temperature coefficient of resonant frequency. The purpose of a ceramic component in a microwave communication system is significantly influenced by its relative permittivity (dielectric constant, (ε_r)) [7]. For resonator, filter and duplexer applications the dielectric constant of the ceramic must be high ($\varepsilon_r > 15$) whereas it should be low ($\varepsilon_r < 10$) for substrate, packaging and antenna applications [8–14]. Ceramic systems having dielectric constant less than 10 facilitate high-speed signal transmission and signal quality by reducing propagation delays, cross talk, noise etc [15–17]. Since the substrate acts as a guided medium for the signal propagation its dielectric loss (tanδ) must be low enough to decrease the signal attenuation or in other words it should hold a high Quality factor (Q) [18–22]. Additionally the ceramic for microwave integrated circuits should hold a near zero temper-

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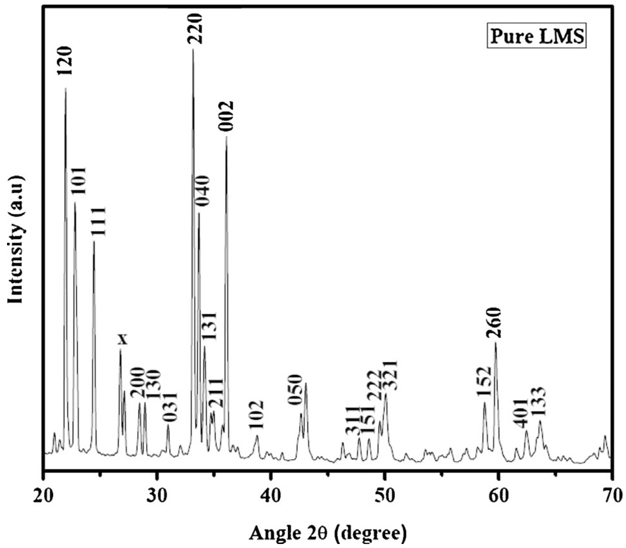
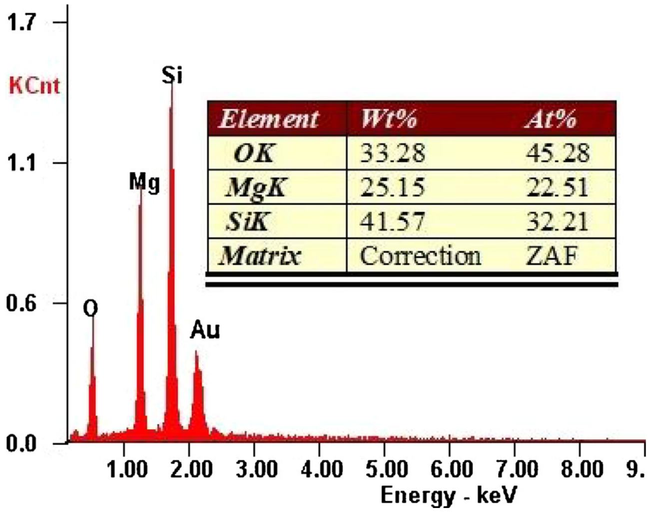


Fig. 1. XRD pattern of LMS.



Element	Wt%	At%
OK	33.28	45.28
MgK	25.15	22.51
SiK	41.57	32.21
Matrix	Correction	ZAF

Fig. 3. EDX spectrum of pure LMS ceramics.

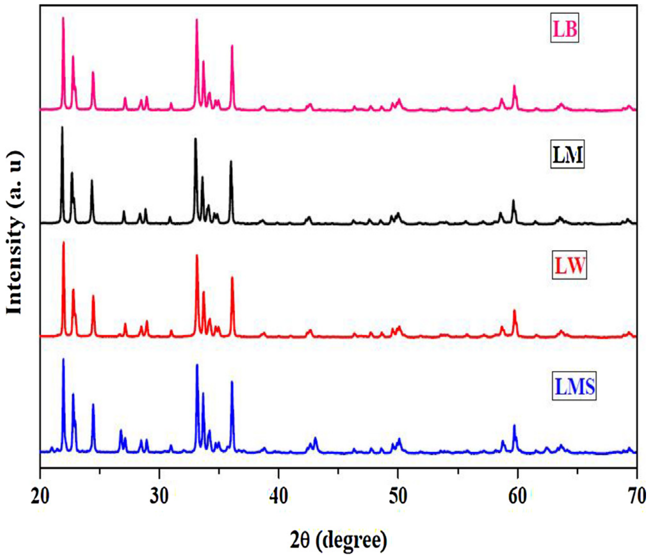


Fig. 2. XRD pattern of LMS with additives (W-WO_3, M-MgF_2, B-B_2O_3).

ature coefficient of resonant frequency for temperature stability [19,23–26].

The majority of microwave ceramics, which show excellent microwave dielectric properties, lack the requisites for substrate applications such as low relative permittivity and low dielectric loss. However quite a few ceramics meeting these requirements have been developed so far [27,28]. Among them phosphates and silicates are suggested to be the potential candidates for high performance substrates [29,30]. Silicates are widely studied as substrate materials because the Si—O bond in SiO_4 tetrahedral owns a covalent nature that leads to low ε_r [29]. For instance Forsterite (Mg_2SiO_4), Zinc Silicate (Zn_2SiO_4), Wollastonite ($CaSiO_3$), Lithium Magnesium Silicate (Li_2MgSiO_4), and numerous rare earth based silicates are reported to have low ε_r and tanδ [31–35]. Even though, a drawback associated with some silicate-based ceramics is their relatively low densification [25]. Since the microwave dielectric property of a ceramic material is extrinsically related to its relative density, high densification must be achieved for substrate applications [36]. Liquid phase sintering which occurs as a result of glass addition aids the densification and lowers the sintering temperature without much distressing the dielectric properties [37]. Thus glass addition is known to be the best and feasible method for enhancing densification of the ceramics. Boron rich glasses such as Boria (Boric Oxide (B_2O_3)), Calcium borosilicate, Lithium borosilicate (LBS) etc are widely investigated since they endorse densification through liquid phase sintering [38–40]. Annrose Sunny et al. analyzed the microwave dielectric properties of novel $LiInSiO_4$ ceramic and successfully modified its properties by the addition of B_2O_3 and Lithium Magnesium Zinc Borosilicate (LMZBS) glasses [7]. Sumesh George et al. reported the synthesis of Li_2MgSiO_4 with the addition of glass compounds such as LBS and LMZBS for LTCC substrate applications [1]. They optimized the sintering temperature and additive weight percentage for which good microwave dielectric properties and densification were revealed. In an article published by Simeon Agathopoulos, the effect of B_2O_3 additive on the sintering temperature, densification, microstructure, and dielectric properties of Li_2MgSiO_4 is examined [41]. Several works based on additives like oxides (ZrO_2, TiO_2, MnO_2, ZnO, CuO etc) and fluorides (LiF, CaF_2) convey the efficiency of these materials in tailoring the properties of microwave ceramics [12,42–48]. Fluorides such as LiF, CaF_2, MgF_2, PbF_2 are low melting point compounds which can promote liquid phase sintering. Thus the fluoride added ceramics can achieve a dense microstructure at lower sintering temperatures. Addition of fluorides endorses the grain growth and decreases the grain boundaries leading to low dielectric loss. On the other hand, glass addition is a widely accepted and established method for reducing sintering temperature of microwave ceramics without declining their dielectric properties. Glasses such as B_2O_3, SiO_2, glass systems including metal oxides and a broad range of glass systems have been studied so far. These additives aid liquid phase sintering and easily form a highly densified microstructure thereby lessening the tangent loss. Oxides can be considered as the best additives for obtaining ceramics with highly ordered microstructure and good dielectric properties at very low weight percentages. For small weight percentages of oxide additives chances of secondary phase formation are very less. Uniform grain size, densification at lower sintering temperatures, and excellent dielectric properties can be simultaneously attained by oxide addition.

In this manuscript, the solid-state synthesis and microwave dielectric properties of Li_2MgSiO_4 and the changes occurred in its properties due to the incorporation of various additives have been presented. These additives namely B_2O_3, MgF_2, and WO_3 were selected from the above-mentioned groups such as glass, fluoride, and oxide respectively. Ceramic samples were synthesized with the

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