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Synthesis and characterisation of novel low temperature ceramic and its implementation as substrate in dual segment CDRA

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ABSTRACT

Li_2O-(2-3x)MgO-(x)Al_2O_3-P_2O_5 (LMAP) (x = 0.00–0.08) ceramic system was prepared through solid state synthesis route at different sintering temperatures (800–925 °C). A small addition of Al_2O_3 (x = 0.02) in LMAP ceramics lowers the sintering temperature by more than 100 °C with good relative density of 94.13%. The sintered samples were characterized in terms of density, apparent porosity, water absorption, crystal structure, micro-structure and microwave dielectric properties. Silver compatibility test is also performed for its use as electrode material in low temperature co-fired ceramic (LTCC) application. To check the performance of the prepared LTCC as substrate, a microstrip-fed aperture-coupled dual segment cylindrical dielectric resonator antenna (DS-CDRA) is designed using LMAP (x = 0.02) ceramic as substrate material and Barium Strontium Titanate with 10 wt% of PbO-BaO-B_2O_3-SiO_2 glass (BSTG) and Teflon as the components of resonating material. The simulation study of the DS-CDRA is performed using the Ansys High Frequency Structure Simulator (HFSS) software. A conductive coating of silver is used on the substrate. The simulated and measured −10 dB reflection coefficient bandwidths of 910 MHz (9.07–9.98 GHz at resonant frequency of 9.49 GHz) and 1080 MHz (8.68–9.76 GHz at resonant frequency of 9.36 GHz), respectively are achieved. The measured results of the fabricated antenna are found in good agreement with the simulation results. The prepared material can find potential applications in radar and radio navigation as well as radio astronomy and military satellite communication.

1. Introduction

Ceramic materials find application as substrates in integrated circuits, thermally stable resonators, filters, oscillators, phase shifters, isolators, circulators and dielectric resonator antennas in microwave and millimeter wave communication and radar systems. Low dielectric constant, low loss tangent and good thermal stability are primary requirements for substrate materials to achieve efficient signal transmission. Low loss ceramic materials used in the circuit improve its overall quality factor by reducing the power dissipation and the insertion loss, which suppresses the electrical noise in oscillators and can produce highly selective filters, low loss phase shifters and non-reciprocal devices [1]. Further, by selecting a low loss ceramic material for realizing a dielectric resonator antenna (DRA), the radiation efficiency can be enhanced due to low conductor and dielectric losses as well as absence of surface waves associated with DRA. Different manufacturers produce similar components for specific applications, however, there are subtle differences in their circuit designs, construction and packaging.

The material requirements for substrate applications are: low dielectric constant ($\varepsilon_r < 20$), Qf > 1000, low temperature variation of dielectric constant (Tk), high thermal conductivity, preferably low thermal expansion, chemical compatibility with the electrode material. Low sintering temperatures <950 °C with good densification are required to avoid melting metallic conductors like silver or gold in the fabrication of ceramic devices. Low temperature co-fired ceramics (LTCC) technology enables the fabrication of three dimensional ceramic modules with embedded silver or gold electrodes [2].

There are many ceramic materials with excellent microwave dielectric properties. However, these materials generally have high sintering temperatures [2]. Low sintering temperature is required for co-firing the ceramic with the electrode material e.g. silver or copper. In recent years, many researchers have found various materials with low sintering temperature (<1000 °C) and matching coefficient of thermal expansion with other co-firing materials for integration with miniaturized microwave

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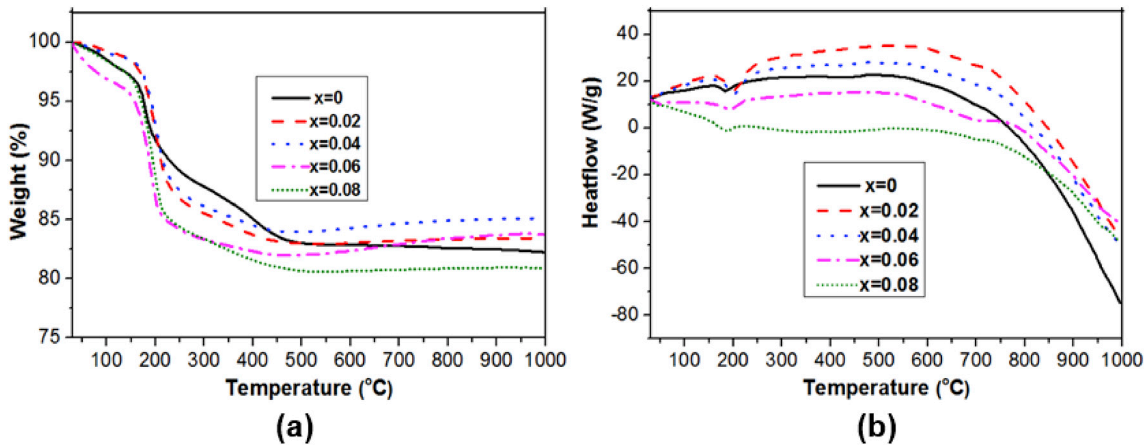

Fig. 1. (a) TG plot and (b) DTA plot of LMAP ceramics.

devices [3]. Also, only a handful of ultra-low temperature co-fired ceramic (ULTTC) materials with sintering temperatures less than 700 °C are developed [4–7]. Recently, Sebastian et al. reported a broad review on the current status of the LTCC [1,2] and ULTCC [8] materials developed so far.

To lower the sintering temperature, generally glass with low softening temperature is added to the ceramic systems. However, the addition of glass increases the loss tangent of the composite due to presence of network formers in the glasses. The microwave energy is possibly absorbed by the network formers [8]. There are two approaches to develop glass based LTCC materials i.e. glass-ceramic and glass + ceramic routes. The densification is achieved through controlled crystallization in glass ceramics [1,2,9]and liquid phase sintering in glass + ceramic routes [10–13] respectively. The commercially available LTCC materials are generally glass-ceramic or glass + ceramic materials. Recently, emphasis is given for the development of glass free ceramic systems to achieve good microwave dielectric properties with low loss and a small number of investigations has been reported for glass free ceramic systems with low sintering temperature [4,6,14–20].

The $LiMgPO_4$ ceramic system has been reported previously as a suitable LTCC material. Many researchers have studied this system by replacing Mg^{2+} with various transition metal elements like Zn, Cu, Ni, Co in different proportions [17–19,21,22]. Investigations on $LiMg_{(1-x)}A_xPO_4$ ceramic (with A = Zn^{+2}, Co^{+2}, Ni^{+2}) shows good microwave dielectric properties and chemical compatibility with electrode material silver. Dong et al. [23]. have studied $Ba_3(VO_4)_2$-$LiMgPO_4$ composite ceramics with good microwave dielectric properties and low sinterability temperature down to 850 °C.

Low sintering temperature material with good microwave dielectric properties can be utilized for microwave device applications like filters, couplers, diplexers, microstrip antennas and dielectric resonator antennas [24–30]. We believe that utilizing the prepared material is a viable approach for substrate application in microwave devices.

Initial part of this paper focuses on the study of the effect of Al_2O_3 addition in Li_2O-(2-3x)MgO-(x)Al_2O_3-P_2O_5 (LMAP) ceramics on their structural and dielectric properties. It is also important to study the design and development of microwave components and DRAs using LTCC substrates. Therefore, the second part of paper describes the study of a DS-CDRA designed using the prepared LMAP (x = 0.02) LTCC ceramic material. The simulation study on the proposed antenna was performed using Ansys HFSS software [31] and the simulation results for the DS-CDRA are compared with the respective measured results.

2. Experiments

2.1. Material synthesis and characterisation techniques

A series of samples having formula in Li_2O-(2-3x)MgO-(x)Al_2O_3-P_2O_5(LMAP), where x = 0, x = 0.02, x = 0.04, x = 0.06 and x = 0.08, were synthesized by conventional solid state route. High purity Li_2CO_3 (99%, Sigma Aldrich), MgO (99%, Sigma-Aldrich), Al_2O_3 (99.5%, Sigma Aldrich) and $NH_4H_2PO_4$ (99%, Alfa Aesar) were used as starting raw materials. Stoichiometric amount of raw materials were mixed thoroughly in ethanol medium for 8 h at 300 rpm using a Fritsch Planetary mill (Pulverisette 5/4). The ball milled batches were dried in oven at 50 °C for 16 h. Thermal reaction and decomposition behaviour of different batch compositions were studied using a Mettler Toledo (STAR system) TGA/DTA machine within the temperature range 30–1000 °C using a heating rate of 5 °C/min in argon atmosphere. The mixed powders were primarily calcined in air at 500 °C for 4 h followed by second calcination at 800 °C for 4 h with intermediate grinding. Calcined powder was ground into fine powder. Powder X-Ray diffraction patterns of the calcined sample were recorded using Rigaku X-ray diffraction with Cu K-Alpha radiation employing a Ni filter with scan rate of 2° per minute. The powders were pressed uniaxially to form cylindrical (diameter = 12 mm, thickness = 2 mm) and rectangular (length = 23 mm, width = 11 mm, height = 2–3 mm) pellets applying a load of 15 MPa. All pellets were sintered in the temperature range 825–925 °C with the heating and cooling rate of 2 °C per minute. The bulk density, water absorption and apparent porosity of the sintered pellets were determined by Archimedes principle. Maximum densification occurs at 825 °C for the LMAP samples with x = 0.02 to 0.08. For the pure sample (x = 0), the densification increases with the increase in sintering temperature up to 925 °C. It was also reported in the previous study that the maximum density for $LiMgPO_4$ ceramic is achieved at 950 °C and above this temperature; the densification decreases further [17]. All the pellets were sintered at optimized sintering temperature of 825 °C for 6 h. The surface morphology and energy dispersive spectra of all the sintered samples were carried out using Scanning Electron Microscope (ZEISS Instruments). High frequency dielectric measurements were carried out through transmission/reflection method within the frequency range 8.2–12.4 GHz using ENA E5071C Keysight Technologies make Network Analyser.

For the fabrication of DS-CDRA, Barium Strontium Titanate (BST) with 10 wt% of PbO-BaO-B_2O_3-SiO_2 glass (BSTG) [32] and teflon were used as the components of resonating material. The square substrate of LMAP (x = 0.02) ceramic having dimensions 42 mm × 42 mm × 1.5 mm was prepared by uniaxially pressing the calcined powder under hydraulic press. After sintering at 825 °C for 6 h, the obtained substrate is polished and made to the dimension of 40 mm × 40 mm × 1.2 mm.

2.2. Design of antenna on LTCC substrate

In order to check the suitability of the prepared material for application in microwave devices, a DS-CDRA antenna was designed and