Available online at www.sciencedirect.com

ScienceDirect

Ceramics International 42 (2016) 5911–5920

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Study on microwave dielectric properties of corundum type $(Mg_{1-x}Co_x)_4Ta_2O_9$ ($x=0-0.6$) ceramics for designing a microwave low pass filter

Parthasarathi Mohanty[a], Sunita Keshri[a,*], Manish Kumar Sinha[a], Vibha Rani Gupta[b]

[a]Department of Physics, Birla Institute of Technology, Mesra, Ranchi 835215, India
[b]Department of Electronics and Communication Engineering, Birla Institute of Technology, Mesra, Ranchi 835215, India

Received 26 November 2015; accepted 23 December 2015
Available online 5 January 2016

Abstract

This paper reports the study of structural, morphological and microwave dielectric properties of $(Mg_{1-x}Co_x)_4Ta_2O_9$ ($x=0$–0.6) ceramics, sintered at 1300–1400 °C. The grown samples were characterized by means of X-ray diffraction, Fourier transform infrared spectroscopy, scanning electron microscopy and energy dispersive X-ray spectroscopy. Microwave dielectric properties of these samples were investigated using the $TE_{01\delta}$ dielectric resonator method. Our study shows that the dielectric properties are strongly dependent on the compositions, densification and micro-structure of the specimens. Out of these compositions, $(Mg_{0.7}Co_{0.3})_4Ta_2O_9$ shows near zero τ_f with a dielectric constant $\epsilon_r \sim 13.79$ and a quality factor $Qf \sim 56{,}876$ GHz (measured at 8.93 GHz). On a substrate made up of this ceramic, a low pass filter of 2.6 GHz cut-off was simulated using Sonnet Lite software and then fabricated. The measured results show a reasonably good agreement with the simulated results. This dielectric shows ~68% size reduction as comparison to FR4 substrate for the same filter design.

Keywords: Corundum; Microwave dielectrics; Temperature coefficient of resonant frequency; Filter circuit

1. Introduction

The development of dielectric resonators for application in microwave communication systems, such as cellular phones, radio frequency integrated circuit (RFIC), global positioning systems (GPS) etc. has been rapidly progressing since past decades [1,2]. Several types of dielectric ceramic materials have been developed and investigated to match with the requirements of dielectric resonators [3]. For such applications, the microwave dielectric ceramics should have high dielectric constant (ε_r) for size miniaturization, large value of quality factor (Qf) for high frequency selectivity and nearly zero temperature coefficient of resonant frequency (τ_f) for thermal stable circuits. Many dielectric ceramics of corundum [4], perovskite [5], double perovskite [6] structures and their composites [1,7] etc. have been searched out for fabrication of different microwave devices. However, improvement of these materials in terms of their desired characteristics is still an important issue for engineering applications.

The dielectric ceramics $A_4M_2O_9$ (where $A = Mg^{2+}$, Co^{2+}, Zn^{2+} and Ni^{2+}; $M = Ta^{5+}$ and Nb^{5+}) with corundum type structure, a crystalline form of aluminium oxide (modified α–Al_2O_3), are found to have wide applications for designing microwave circuits as these possess high quality factor and low dielectric loss [2,8–11]. These ceramics have achieved much attention as they exhibit rich structural and physical properties. Out of different corundum structured ceramics, $Mg_4Ta_2O_9$ has attracted much interest because of its high Qf value and low dielectric loss [2]. In this compound, each TaO_6 octahedral layer is sandwiched by two MgO_6 octahedron layers being a modified α–Al_2O_3 structure as found in case of $Mg_4Nb_2O_9$ [9]. This compound has a trigonal crystal structure and space group

*Corresponding author.
E-mail addresses: s_keshri@bitmesra.ac.in, sskeshri@rediffmail.com (S. Keshri).

http://dx.doi.org/10.1016/j.ceramint.2015.12.139

of $P\overline{3}c1$. Mei et al. [2] have reported the microwave dielectric properties of $Mg_4Ta_2O_9$ as $\varepsilon_r = 11.9$, $Qf = 195,000$ GHz and $\tau_f = -47$ ppm/°C. But such a large non zero value of τ_f creates a serious problem for microwave communication. In case of B-site substitution of V^{5+} in $Mg_4(TaNb_{1-x}V_x)O_9$ compound, Kan et al. [11] have obtained high Qf value ($\sim$200,000 GHz) but a large and negative τ_f(~ -73 ppm/°C). Similarly, Ogawa et al. [12] have found the τ_f value between -60 and –70 ppm/°C and Qf value $\sim$350,000 GHz when Ta is substituted in B-site of $Mg_4(Nb_{2-x}Ta_x)O_9$. However, Zhang et al. [13] have improvised τ_f value to zero by A-site substitution of La^{3+} in $(Nd_{1-x}La_x)_{1.02}Nb_{0.988}O_4$. Literature survey reveals that with partial replacement of Mg^{2+} by Zn^{2+} in $Mg_4Ta_2O_9$ shows a much higher Qf value than that of the parent sample, but with non zero τ_f (~ -67 ppm/°C) [14]. However, to the best of our knowledge, in case of this ceramic no attempt has been made yet to replace Mg by Co.

In this paper the structural and the microwave dielectric properties of $(Mg_{1-x}Co_x)_4Ta_2O_9$ ($x = 0$–0.6) ceramics, sintered at 1300–1400 °C have been investigated using X-ray diffractometer (XRD), scanning electron microscope (SEM) and Fourier transform infrared (FTIR) spectroscope. The $x = 0.3$ sample of this series has shown thermal stability ($\tau_f \sim 0$) in microwave range. To show the advantage of this material a low pass Chevyshev-type stepped impedance microstrip filter has been fabricated. Full wave EM performance of this filter has been compared with the simulated result obtained using Sonnet Lite software.

2. Experimental details

The ceramic samples of $(Mg_{1-x}Co_x)_4Ta_2O_9$ ($x = 0$–0.6) were synthesized using a solid state mixed oxide route with starting materials of high-purity oxide powders ($\sim$ 99.9%): MgO, Co_3O_4 and Ta_2O_5. These powders were taken as per the desired stoichiometry. Weighted raw materials were ground in agate mortar and pestle for 6 h and then calcined for 4 h. A series of cylindrical pellet like samples were prepared using a uniaxial hydraulic press at 1000 kg/cm^2. These pellets were sintered at 1300–1400 °C for 6 h with heating rate of 5 °C/min in air.

The crystalline behaviour of the sintered ceramics was studied by X-ray diffraction using Bruker D–8 advance diffractometer with CuK_α ($\lambda = 1.5406$ Å) as X-ray source for the range $20° \leq 2\theta \leq 80°$ taking step size 0.02°. The lattice parameters were thoroughly investigated using Checkcell software. The FTIR spectra for different modes were observed using IR spectroscopy (IR Prestige–21). The topography study and elemental analysis of the prepared samples were carried out using JEOL–6330F SEM equipped with Oxford INCA energy dispersive X-ray (EDX) spectrometer.

The apparent density as well as apparent porosity of the grown samples was calculated using Archimedes principle. The dry weight (D) of the sintered sample was measured and a glass beaker with the sample immersed in distilled water was kept in a vacuum chamber for 3 h, then the beaker was taken out from the chamber. Pores present in the pellet were now completely filled with water and the soaked weight (W) of the pellet was noted. Suspended weight (I) was taken by hanging the pellet inside the water. Apparent density ($\rho_{apparent}$) along with the apparent porosity were measured using following relations of Archimedes principle:

$$\rho_{apparent} = \frac{D}{W - I} \quad (1)$$

$$\text{Apparent porosity} = \frac{W - D}{W - I} \quad (2)$$

The theoretical density of the sample was calculated using,

$$\rho_{theory} = \frac{ZA}{VN_A}, \quad (3)$$

where Z represents the number of atoms per unit cell, A represents atomic weight (g/mol), V, the volume of the unit cell (cm^3) and N_A stands for Avogadro number (mol^{-1}). To calculate the relative density the following relation was used:

$$\rho_{relative} = \frac{\rho_{apparent}}{\rho_{theory}} \quad (4)$$

The dielectric constant, quality factor and temperature coefficient of resonant frequency at microwave range were measured using $TE_{01\delta}$ resonance mode of the cylindrical pellets inserted in a shielding cavity by Agilent PNA N5230A network analyser. According to Hakki and Coleman [15], the dielectric constant can be calculated from the following equation:

$$\varepsilon_r = 1 + \frac{c}{\pi d f_0}\left(\alpha_1^2 + \beta_1^2\right), \quad (5)$$

where c is the velocity of light, α_1 can be obtained by the mode chart [16] and β_1 can be obtained from the resonant frequency (f_0) and the sample dimension. Here d represents the diameter of the cylindrical pellet. On the basis of electro-dynamical analysis, the dielectric parameters were calculated using the observed resonant frequency and structural dimensions of pellets. Keeping the pellet in a temperature controlled cylindrical cavity, the temperature coefficient of resonant frequency (τ_f) of the sample was measured by the following equation

$$\tau_f = \frac{f_2 - f_1}{f_1(T_2 - T_1)}, \quad (6)$$

where f_1 and f_2 represent the resonant frequencies at two different temperatures T_1 (30 °C) and T_2 (80 °C), respectively.

To verify the advantages of these ceramics, a low pass filter of 2.6 GHz cut-off frequency was simulated and fabricated on a substrate made by one of the grown samples, $(Mg_{0.7}Co_{0.3})_4Ta_2O_9$ having $\tau_f \sim 0$. Simulation was done using Sonnet Lite software. The performance of this filter was compared with a filter of similar design fabricated on FR4 substrate.