Preparation and microwave dielectric properties of Li$_3$(Mg$_{0.92}$Zn$_{0.08}$)$_2$NbO$_6$–Ba$_3$(VO$_4$)$_2$ composite ceramics for LTCC applications

Tianwen Zhang, Ruzhong Zuo, Chen Zhang

Institute of Electro Ceramics & Devices, School of Materials Science and Engineering, Hefei University of Technology, Hefei 230009, PR China

A R T I C L E   I N F O

Article history:
Received 25 July 2014
Received in revised form 24 February 2015
Accepted 7 March 2015
Available online 10 March 2015

Keywords:
A. Ceramics
A. Composites
C. X-ray diffraction
D. Dielectric properties

A B S T R A C T

In this work, the (1–x)Li$_3$(Mg$_{0.92}$Zn$_{0.08}$)$_2$NbO$_6$–xBa$_3$(VO$_4$)$_2$ (x = 0.1–0.35) ceramics were prepared via a conventional solid state reaction route. The phase composition, microstructure and microwave dielectric properties were investigated by an X-ray diffractometer (XRD), a scanning electron microscope and a network analyzer. The XRD results indicated that the Li$_3$(Mg$_{0.92}$Zn$_{0.08}$)$_2$NbO$_6$ and Ba$_3$(VO$_4$)$_2$ phases could well coexist without forming any secondary phases. The dielectric constant (e$_r$) and quality factor (Q×f) values of the Li$_3$(Mg$_{0.92}$Zn$_{0.08}$)$_2$NbO$_6$ ceramic decreased with the addition of Ba$_3$(VO$_4$)$_2$ phase, however its temperature coefficient of resonant frequency (τ$_r$) value was improved significantly. Excellent microwave dielectric properties of e$_r$ ~ 16.3, Q×f ~ 50,084 GHz (at 8.64 GHz) and τ$_r$ ~ 15 ppm/°C were achieved for the x = 0.3 sample when sintered at 950 °C for 4 h. The chemical compatibility with Ag electrode indicated that the 0.7Li$_3$(Mg$_{0.92}$Zn$_{0.08}$)$_2$NbO$_6$–0.3Ba$_3$(VO$_4$)$_2$, composite ceramic would be a promising material for the low temperature co-fired ceramic applications.

© 2015 Elsevier Ltd. All rights reserved.

1. Introduction

With the rapid development of modern microwave communication systems, microwave dielectric ceramics have attracted much scientific and commercial attention. These materials should have a proper dielectric constant (e$_r$) (that is to say, a low e$_r$ is useful for reducing the signal delay, while a high e$_r$ is desirable for device miniaturization), a high quality factor (Q) for improving the signal-to-noise ratio, and a near-zero temperature coefficient of resonant frequency (τ$_r$) (~10 ppm/°C ≤ τ$_r$ ≤ +10 ppm/°C) for the stability of the transmitted frequency [1,2]. The above three parameters are the key requirements for industrial production and practical application in addition to the low cost owing to cheaper raw materials and lower sintering temperatures.

So far, various dielectric ceramics with a low e$_r$ and a high Q have been studied, such as silicate (Mg$_2$SiO$_4$, Zn$_2$SiO$_4$) [3,4], aluminate (ZnAl$_2$O$_4$, MgAl$_2$O$_4$) [5,6], niobate (MgNb$_2$O$_6$, Mg$_3$Nb$_2$O$_9$) [7], and titanate (MgTiO$_3$, Mg$_{0.95}$Zn$_{0.05}$TiO$_3$) [8,9]. Their higher Q values are advantageous compared with other materials, but the large negative τ$_r$ values limit their further applications in microwave devices. A lot of research work has been focused on various processing techniques to adjust the τ$_r$ value to near zero. One way is to combine two components with opposite τ$_r$ values, which has been employed widely because of an obvious effect, such as in MgAl$_2$O$_4$–TiO$_2$ [6], MgTiO$_3$–CaTiO$_3$ [8] and (Mg$_{0.95}$Zn$_{0.05}$TiO$_3$)–SrTiO$_3$ [9]. The other way is to change the bond valences of A- or B-sites or induce the tilting of oxygen octahedra by means of ionic substitution. However, the impact of the latter was generally not notable and could be usually found in some individual structures such as complex perovskites [10] and tetragonal scheelite [11].

It is of particular note that Ba$_3$(VO$_4$)$_2$ (BV) not only owns a positive τ$_r$ value (τ$_r$ ~ +52 ppm/°C) and a relatively low sintering temperature ~1100 °C, but also possesses good microwave properties of e$_r$ ~ 14 and Q×f ~ 42,000 GHz [12]. It has been employed as a positive τ$_r$-tailoring material in many composite ceramics, for instance BV–LiMg$_{0.5}$Zn$_{0.5}$PO$_4$, BV–CaWO$_4$, BV–Zn$_{0.9}$SiO$_4$–x, BV–MgSiO$_4$ [13–16], in which the sintering temperature could be lowered obviously in addition to acceptable microwave dielectric properties. In these composite ceramics, the two components often form diphasic compositions instead of solid solutions because of their different crystal structures. As a result, the variation of microwave dielectric properties could be explained by a mixing rule. The Li$_3$(Mg$_{0.92}$Zn$_{0.08}$)$_2$NbO$_6$ (LMZN) ceramic has been reported to own excellent microwave properties of e$_r$ ~ 17.2, Q×f ~ 142,331 GHz and τ$_r$ ~ 24.1 ppm/°C in our...