Structure and microwave dielectric properties of Ba$_{1-x}$Sr$_x$Mg$_2$V$_2$O$_8$ ceramics

Yang Wang, Ruzhong Zuo *

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

ARTICLE INFO

Article history:
Received 14 March 2016
Received in revised form 23 March 2016
Accepted 28 March 2016
Available online 29 March 2016

Keywords:
C. Dielectric properties
Ceramics
Bond valence

ABSTRACT

The Ba$_{1-x}$Sr$_x$Mg$_2$V$_2$O$_8$ ($0 \leq x \leq 0.4$) microwave dielectric ceramics were fabricated by a standard solid-state reaction method. The formation of a continuous solid solution within the whole composition range was identified. The ceramic samples could be well densified in the temperature range of 885–975 °C in air for 4 h. The permittivity ($\varepsilon_r$) was found to increase with increasing ionic polarizabilities. The Q × f values were believed to be closely related with packing fraction and grain refinement. The Sr$^{2+}$ substitution contributed to a monotonous increase of the A-site bond valence, such that the $\tau_f$ value experienced a considerable variation from negative to positive values. The optimum microwave dielectric properties of an $\varepsilon_r$ of 13.3, a high Qxf of 86.640 GHz (9.6 Hz) and a near-zero $\tau_f$ of −6 ppm/°C could be yielded in the $x=0.15$ sample when sintered at 915 °C for 4 h.

1. Introduction

A rapid development in wireless communication systems such as satellite positioning, mobile phones and patch antennas has brought demands for high-performance microwave devices over the past decades [1]. Moreover, the low-temperature cofired ceramic (LTCC) technology for the miniaturization and integration of electronic devices has generally required the corresponding dielectric materials to have a lower sintering temperature than the melting point of the Ag electrode, in addition to an appropriate permittivity ($\varepsilon_r$), a high quality factor (Qxf) value, and a near-zero temperature coefficient of resonant frequency ($\tau_f$) [2–3]. Most of microwave dielectric materials need to be densified at higher sintering temperatures [4–7]. This situation can be improved through the addition of low melting glass, chemical processing or use of fine raw materials [8–10]. However, the complicated fabrication processing and deteriorated microwave dielectric properties have become challenges for the application of these LTCC materials. A glass-free LTCC material with desirable microwave dielectric properties is therefore of much interest. A lot of efforts have been recently focused on some Li$_2$O–TeO$_2$–Bi$_2$O$_3$–MoO$_3$– and V$_2$O$_5$–based compounds with intrinsically low sintering temperatures and desirable performances [11–20]. For example, some vanadates with a garnet structure were reported to show great potentials in LTCC applications [21–23].

The BaMg$_2$V$_2$O$_8$ ceramic with a body-centered tetragonal structure demonstrated excellent microwave dielectric properties of $\varepsilon_r=12$, $Q\times f=156$, 140 GHz (9.9 GHz) and $\tau_f=-36$ ppm/°C as sintered at a relatively low temperature of 900 °C [24]. The low-firing characteristics and ultra-low loss probably make it be a promising candidate for advanced substrate materials used in microwave integrated circuits. However, the negative $\tau_f$ value might strongly restrict its application in LTCC-based devices. A BaMg$_2$V$_2$O$_8$–TiO$_2$ composite was prepared in our previous work using two materials with opposite signs of $\tau_f$. This approach can well explain the variation of $\tau_f$ values form the viewpoint of the two-phase coexistence. In addition, the A-site ionic substitution has been widely attempted owing to its role in tailoring the undesirable $\tau_f$ value of the matrix by forming a solid solution. It was previously revealed that the substitution of smaller Ni ions (0.69 Å, CN=6) for Zn ions (0.74 Å, CN=6) contributed to the decrease of the A-site bond valence and consequently the decrease of $\tau_f$ values in the monoclinic structured (Zn$_{1-x}$Ni$_x$)$_2$Nb$_2$O$_7$ system [25]. Moreover, the increase of the oxygen bond valence in tetragonal scheelite compounds was reported to result in the increase of the average bond strength of A-site and B-site ions, which in turn enhanced the restoring force for the tilting of A$_2$BO tetrahedra, such that the $\tau_f$ value decreased [26]. For perovskite solid solutions, the $\tau_f$ values are usually affected by either the variation of polarizability per unit cell or a phase transition associated with octahedral rotations [27–29]. The substitution of smaller A-site ions for Ba$^{2+}$ in Ba$_{3}$MM$_2$O$_9$ (M=Mg, Ni; Zn; M′=Nb, Ta) perovskites was believed to contribute to the change of octahedral tilting, therefore producing a corresponding adjustment of $\tau_f$ [30].

In current work, Sr$^{2+}$ was anticipated to occupy the A-site in the BaMg$_2$V$_2$O$_8$ lattice within the entire composition range. The