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

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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 ε_r was found to increase with increasing ionic polarizabilities. The $Q \times f$ 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 τ_f value experienced a considerable variation from negative to positive values. The optimum microwave dielectric properties of an ε_r of 13.3, a high Qxf of 86,640 GHz (9.6 Hz) and a near-zero τ_f of -6 ppm/°C could be yielded in the $x=0.15$ sample when sintered at 915 °C for 4 h.

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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 (ε_r), a high quality factor (Qxf) value, and a near-zero temperature coefficient of resonant frequency (τ_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 τ_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 τ_f. This approach can well explain the variation of τ_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 τ_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 τ_f values in the monoclinic structured (Zn$_{1-x}$Ni$_x$)$_3$Nb$_2$O$_8$ 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 τ_f value decreased [26]. For perovskite solid solutions, the τ_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 τ_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