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Microwave dielectric properties of $Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O_3$ ceramics

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Abstract

The microwave dielectric properties of ceramics based on $Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O_3$ is investigated as a function of x. The densification as well as dielectric properties deteriorate with increase in the substitution levels of $(Ti_{1/3}W_{1/3})^{3.33+}$ at $(Ta_{2/3})^{3.33+}$ site in $Ba(Mg_{1/3}Ta_{2/3})O_3$. The τ_f is approaching zero between $x = 0.1$ and 0.15 in $Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O_3$ where quality factor is reasonably good ($Q_u \times f = 80{,}000$–$90{,}000$ GHz). The $Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O_3$ with $x = 1.0$ has $\varepsilon_r = 15.4$, $\tau_f = -25.1$ ppm/°C, $Q_u \times f = 35{,}400$ GHz.

Keywords: A. Ceramics; A. Oxides; C. X-ray diffraction; D. Crystal structure; D. Dielectric properties

1. Introduction

The complex perovskites $Ba(Mg_{1/3}Ta_{2/3})O_3$ [BMT] shows very interesting dielectric properties in the microwave frequency region [1]. The crystal chemistry of A-site and B-site substitution in Ba- and Pb-based 1:2 ordered complex perovskites are reported [2] to have tremendous influence on its physical and dielectric properties. The cation ordering kinetics in Ba- and Pb-based ceramics are markedly different as the latter disorders even at very low-temperatures [3]. The partial substitution of Ba with Sr in 1:2 ordered complex perovskite niobates have been investigated by a number of investigators for possible applications in microwave communication devices [4]. There have been many attempts [5,6] to predict the microwave dielectric properties by studying the solid solution phases in $Ba_{1-x}Sr_x(Mg_{1/3}Ta_{2/3})O_3$ system, which provided valuable information about the structure–property relation of low-loss complex perovskites. The barium in complex perovskites can be easily substituted with strontium ion as they are isovalent and have comparable ionic radii (1.61 and 1.44 Å, respectively), which are the two important requirements for solid solution formation. The complete replacement of Ba with Ca in the A-site of BMT was undertaken by Kagata and Kato [7] who found a high quality factor of $Q_u \times f = 78{,}000$ GHz for $Ca(Mg_{1/3}Ta_{2/3})O_3$ ceramics. The substitution of a trivalent ion (La) in A-site of BMT results in the coexistence of both 1:1 and 1:2 ordered domains [8,9]. The substitution at the B-site in Ba-based complex perovskites with tetravalent ion has been one of the most interesting part of the research on complex perovskites as this imparts significant effects on the structural order of these materials [10].

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The microwave dielectric properties of $Ba(Mg_{1/3}Ta_{2/3})O_3$–$A(Mg_{1/2}W_{1/2})O_3$ [A = Ba, Sr and Ca] have been investigated by Furuya and Ochi [11] who found that the presence of $Ba(Mg_{1/2}W_{1/2})O_3$ reduces the temperature coefficient of resonant frequency for BMT. The duo could develop a zero τ_f composition for $0.95Ba(Mg_{1/3}Ta_{2/3})O_3$–$0.05Ba(Mg_{1/2}W_{1/2})O_3$ where the $Q_u \times f$ value is reaching as high as 40,000 GHz. A significant attempt in this direction was done by Takahashi et al. [12] who while investigating the microwave dielectric properties of $(1-x)Ba(Mg_{1/2}W_{1/2})O_3$–$xBaTiO_3$ ceramics, found that for $x = 2/3$, the ceramics form a single phase (i.e. $Ba(Mg_{1/3}Ti_{1/3}W_{1/3})O_3$) with low-dielectric constant and high quality factor. The above said composition is corresponding to $x = 1$ in the $Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O_3$ which in principle, is nothing, but a solid solution between $Ta_{2/3}$ and $W_{1/3}Ti_{1/3}$ in BMT. Even though, barium magnesium tantalate and barium zinc tantalate have attractive low-loss properties and are extensively being used in microwave devices, the extremely high cost of the contributing raw material tantalum pentoxide make them less attractive from an economic perspective. Hence, the searches for alternate low-loss materials which are free of tantalum are being sought for active devices in telecommunication industry. So far no useful work has been done on the aspect of simultaneous substitutional characteristics of W and Ti on the Ta site in BMT which has been undertaken in this study. The present paper describes the variation of bulk density and microwave dielectric properties of $Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O_3$ system as a function of x.

2. Experimental

The $Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O_3$ [x = 0.01, 0.02, 0.03, 0.04, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9 and 1.0] ceramics were prepared by the conventional mixed oxide route. High purity (>99.9%) powders of $BaCO_3$, TiO_2, WO_3, $(MgCO_3)_4Mg(OH)_2\cdot 5H_2O$ (Aldrich Chemicals) and Ta_2O_5 (Nuclear Fuel Complex, Hyderabad) were used as the starting materials. They were weighed according to the stoichiometric compositions based on $Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O_3$ [x = 0.0–1.0] and were ball milled in a plastic bottle using zirconia balls in deionized water for 24 h. The slurry was dried in an oven at 100 °C and calcined in platinum crucible at 1200 °C for 10 h with intermediate grinding. The calcined powder was then ground for 2 h and 4 wt.% aqueous solution of PVA was added to it as a binder. The powder was uniaxially pressed into cylindrical compacts of 14 mm diameter and 6–8 mm thickness under a pressure of 150 MPa in tungsten carbide die. These compacts were fired at a rate of 5 °C/min up to 600 °C and soaked at 600 °C for 1 h to expel the binder before they were sintered in the temperature range 1500–1600 °C for 4 h in air at a heating rate of 10 °C/h. The sintered samples were then cooled to 800 °C at a slow rate of 60 °C/h and subsequently annealed at 1350 °C for 20 h. The polished ceramic pellets with an aspect ratio (diameter to height) of 1.8–2.2 which is ideal for maximum separation of the modes, were used for microwave measurements. The bulk density of the sintered samples was measured using Archimedes method. The powdered samples were used for analysing the X-ray diffraction patterns using Cu Kα radiation (Philips X-ray

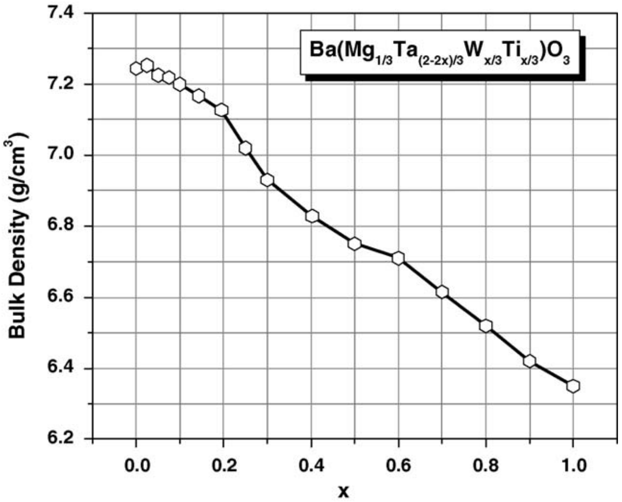

Fig. 1. Variation of bulk density of solid solution phases in $Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O_3$ as a function of x.