



ELSEVIER

Available online at www.sciencedirect.com

SCIENCE @ DIRECT®

**Materials
Research
Bulletin**

Materials Research Bulletin xxx (2005) xxx–xxx

www.elsevier.com/locate/matresbu

Microwave dielectric properties of Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O₃ ceramics

K.P. Surendran^a, P. Mohanan^b, M.T. Sebastian^{a,*}^a Ceramic Technology Division, Regional Research Laboratory, Trivandrum 695 019, India^b Department of Electronics, Cochin University of Science and Technology, Cochin 682 022, India

Received 21 June 2005; received in revised form 6 September 2005; accepted 10 October 2005

Abstract

The microwave dielectric properties of ceramics based on Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O₃ 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})O₃. 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})O₃ 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})O₃ with $x = 1.0$ has $\epsilon_r = 15.4$, $\tau_f = -25.1$ ppm/°C, $Q_u \times f = 35,400$ GHz.

© 2005 Published by Elsevier Ltd.

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})O₃ [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 perovskites 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})O₃ 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})O₃ 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].

* Corresponding author. Tel.: +91 471 2515294; fax: +91 471 2491712.

E-mail address: mailadils@yahoo.com (M.T. Sebastian).

The microwave dielectric properties of $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3\text{-A}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3$ [A = Ba, Sr and Ca] have been investigated by Furuya and Ochi [11] who found that the presence of $\text{Ba}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3$ reduces the temperature coefficient of resonant frequency for BMT. The duo could develop a zero τ_f composition for $0.95\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3\text{-}0.05\text{Ba}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3$ 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 $(1-x)\text{Ba}(\text{Mg}_{1/2}\text{W}_{1/2})\text{O}_3\text{-}x\text{BaTiO}_3$ ceramics, found that for $x = 2/3$, the ceramics form a single phase (i.e. $\text{Ba}(\text{Mg}_{1/3}\text{Ti}_{1/3}\text{W}_{1/3})\text{O}_3$) with low-dielectric constant and high quality factor. The above said composition is corresponding to $x = 1$ in the $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ which in principle, is nothing, but a solid solution between $\text{Ta}_{2/3}$ and $\text{W}_{1/3}\text{Ti}_{1/3}$ 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 $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ system as a function of x .

2. Experimental

The $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ [$x = 0.01, 0.02, 0.03, 0.04, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9$ and 1.0] ceramics were prepared by the conventional mixed oxide route. High purity (>99.9%) powders of BaCO_3 , TiO_2 , WO_3 , $(\text{MgCO}_3)_4\text{Mg}(\text{OH})_2 \cdot 5\text{H}_2\text{O}$ (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 $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ [$x = 0.0\text{--}1.0$] 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 $5^\circ\text{C}/\text{min}$ up to 600°C and soaked at 600°C for 1 h to expel the binder before they were sintered in the temperature range $1500\text{--}1600^\circ\text{C}$ for 4 h in air at a heating rate of $10^\circ\text{C}/\text{h}$. The sintered samples were then cooled to 800°C at a slow rate of $60^\circ\text{C}/\text{h}$ 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 $\text{Cu K}\alpha$ radiation (Philips X-ray

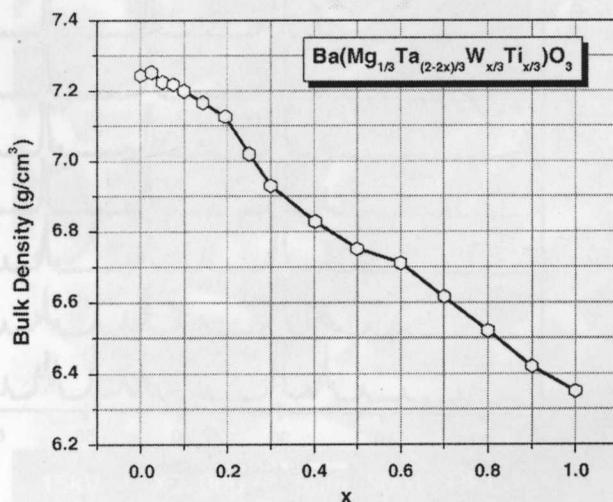


Fig. 1. Variation of bulk density of solid solution phases in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ as a function of x .

69

70 Diffractometer). The sintered samples were thermally etched for 30 min at a temperature of about 25 °C below the
 71 sintering temperature and the surface morphology was studied using a scanning electron microscope (JEOL-JSM
 72 5600 LV, Tokyo, Japan).

73 The dielectric properties of the materials were measured in the microwave frequency range using a network
 74 analyser HP 8510 C (Hewlett-Packard, Palo Alto, CA). The dielectric constant ϵ_r was measured by the post resonator
 75 method of Hakki and Coleman [13] and the samples were end shorted with finely polished copper plates coated with
 76 gold. The microwave is coupled through E-field probes as described by Courtney [14]. TE₀₁₁ mode of resonance
 77 whose Q is intimately related to the dielectric loss ($\tan \delta$), is used for the measurements. The unloaded quality factor
 78 Q_u of the resonance was determined using a copper cavity whose interior was coated with silver and the ceramic
 79 dielectric is placed on a low-loss quartz spacer which reduces effect of losses due to the surface resistivity of the cavity
 80 [15]. For measuring the Q factor, the cavity method is ideal since the electric field is symmetrical with geometry of the
 81 cavity and the dielectric [16]. The coefficient of thermal variation of resonant frequency (τ_f) was measured by noting
 82 the temperature variation of the resonant frequency of TE₀₁₁ mode in the reflection configuration over a range of
 83 temperature 20–80 °C when the sample was kept in the end shorted position.

84 3. Results and discussion

84

85 The densification behavior of Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O₃ phases is plotted in Fig. 1. Using the powder
 86 diffraction method, the theoretical density of BMT is calculated as 7.625 g/cm³ while that of Ba(Mg_{1/3}W_{1/3}Ti_{1/3})O₃

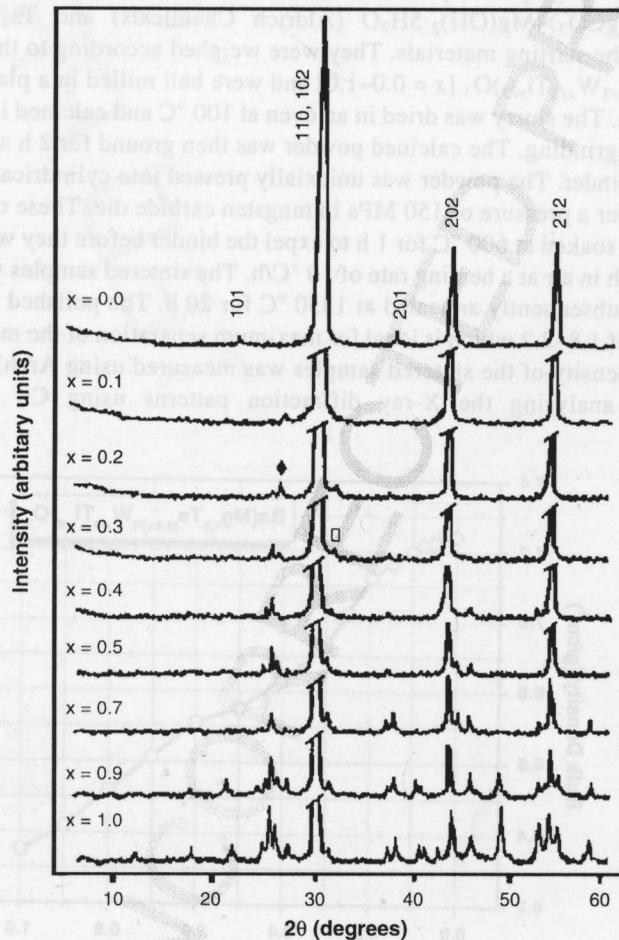


Fig. 2. The powder diffraction pattern of Ba(Mg_{1/3}Ta_{(2-2x)/3}W_{x/3}Ti_{x/3})O₃ [$x = 0.1, 0.2, 0.3, 0.4, 0.5, 0.7, 0.9$ and 1.0]: (◆) represents TiO₂, (□) BaTiO₃.

Table 1

The Miller indices of major powder diffraction profiles for $x = 1.0$ in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ using a hexagonal superstructure of dimensions $a = 5.79(1)$ and $c = 14.16(2)$ Å

Peak no.	2θ (°)	d -Spacing (Å)	hkl
1	12.43	7.121	002
2	17.67	5.018	100
3	22.12	4.018	102
4	25.17	3.538	004
5	25.81	3.453	103
6	27.44	3.250	011
7	28.12	3.173	113
8	30.67	2.915	110
9	31.10	2.876	220
10	37.11	2.423	113
11	38.23	2.354	202
12	40.11	2.248	114
13	41.07	2.197	203
14	43.20	2.094	121
15	44.12	2.053	204
16	47.76	1.906	205
18	49.83	1.829	212
19	53.71	1.706	213
20	54.62	1.680	300
21	55.90	1.645	301
22	58.43	1.579	215

[17] is 6.412 g/cm^3 . As expected, the bulk density of compositions in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ decreases with x .

Fig. 2 represents the powder diffraction pattern of $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ [$x = 0.1, 0.2, 0.3, 0.4, 0.5, 0.7, 0.9$ and 1.0]. It is worthwhile to note that the low-loss ceramic system involving $\text{BaO}-x\text{MgO}-y\text{WO}_3-z\text{TiO}_2$ [$x + y + z = 1$] has already been investigated by Takahashi et al. [18]. They suggested a 1:1:1 cation ordering between $\text{Mg}-\text{W}-\text{Ti}$ along $\langle 100 \rangle$ direction for $x = 0.32, y = 0.32$ and $z = 0.68$ in $\text{BaO}-x\text{MgO}-y\text{WO}_3-z\text{TiO}_2$. They attempted to index the XRD reflections based on a hexagonal unit cell ($a \sim 11.3 \text{ Å} = 2 \times \sqrt{2} \times 4 \text{ Å}$; $c \sim 13.9 \text{ Å} = 2 \times \sqrt{3} \times 4 \text{ Å}$)

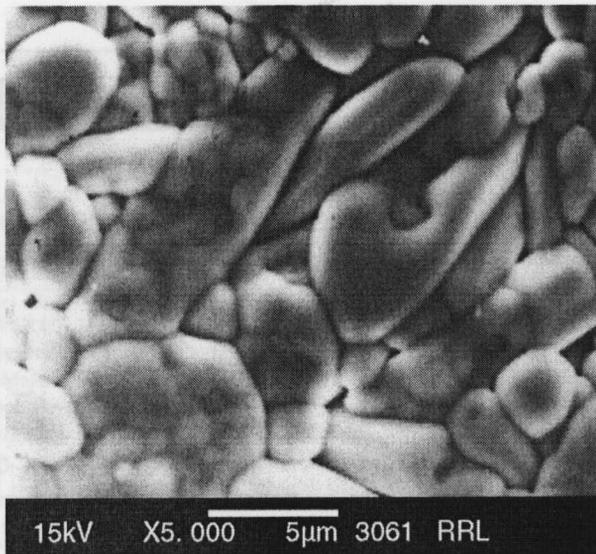


Fig. 3. SEM picture of $\text{Ba}(\text{Mg}_{0.333}\text{Ta}_{0.633}\text{Ti}_{0.016}\text{W}_{0.016})\text{O}_3$.

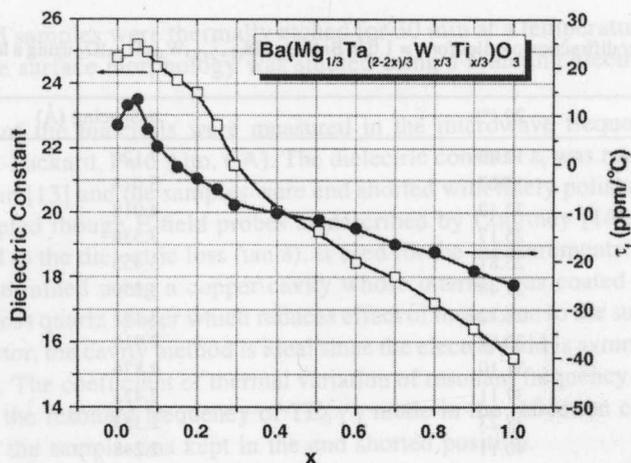


Fig. 4. Variation of the dielectric constant of $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ as a function of x .

93

94 assuming a lattice constant $a \sim 4 \text{ \AA}$ for the pseudo cubic unit cell [19]. In the present investigation also, the indexing of
 95 the XRD pattern is done assuming a hexagonal unit cell with cell parameters $a = 5.79(1)$ and $c = 14.16(2) \text{ \AA}$. Using the
 96 above assumption, the Miller indices for major reflections (for $10 \leq 2\theta \leq 60$) of the Ti + W rich end composition is
 97 given in Table 1. However, based only on powder diffraction analysis, it may be erroneous to arrive at a conclusion
 98 about the formation of an ordered phase with the composition $\text{Ba}(\text{Mg}_{1/3}\text{Ti}_{1/3}\text{W}_{1/3})\text{O}_3$ which corresponds to $x = 1.0$ in
 99 $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ system. A more accurate structural analysis using electron microscopic and
 100 spectroscopic tools is required to establish the 1:1:1 cation ordering between Mg–W–Ti along $\langle 100 \rangle$ direction in
 101 $\text{Ba}(\text{Mg}_{1/3}\text{Ti}_{1/3}\text{W}_{1/3})\text{O}_3$, which is beyond the purview of this investigation. Also it is evident from Fig. 2 that when the
 102 concentration levels of Ti + W increases, presence of trace amount of unreacted TiO_2 (JCPDS File Card Number 21-
 103 1276) was detected in the powder diffraction pattern (see Fig. 2). It is evident from the powder diffraction patterns that
 104 the formation of trace amount of BaTiO_3 (JCPDS File Card Number 24-129) also detected for $0.5 \leq x \leq 0.9$.

105 Fig. 3 represents the scanning electron micrographs recorded from a typical samples for $x = 0.05$, in $\text{Ba}(\text{Mg}_{1/3}$
 106 $\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$. This ceramic corresponds to the highest quality factor among all the compositions studied in
 107 the present report and its average grain size is less than $5 \mu\text{m}$. Since, no additional phases were visible in the SEM, one
 108 can assume that addition of smaller percentages of W + Ti could have resulted in efficient substitution at the Ta site.

109

110

111

112

113

114

115

116

117

118

119

120

121

122

123

124

125

126

127

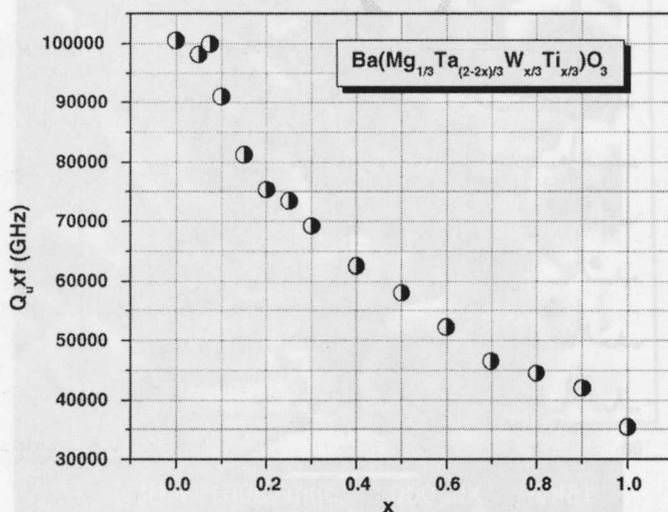


Fig. 5. Variation of the unloaded quality factor of $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ as a function of x .

Fig. 4 is a plot describing the variation of the dielectric constant of the specimens with respect to the compositional variation of W + Ti against Ta. The measured values of dielectric constants were corrected for porosity [20]. For small values of x , the dielectric constant slightly increases (from 24.8 for $x = 0.0$, to 25.21 for $x = 0.05$ in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$). Thereafter, ϵ_r decreases monotonically for higher values of x , reaching, 15.4 for $x = 1.0$ in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$. This dielectric constant for the end composition, is in good agreement with a previous report by Takahashi et al. [18] who measured the ϵ_r of $\text{Ba}(\text{Mg}_{1/3}\text{Ti}_{1/3}\text{W}_{1/3})\text{O}_3$ as ~ 14 .

The temperature coefficient of resonant frequency (τ_f) of the compositions in studied in the present investigation is plotted in Fig. 4. The τ_f of stoichiometric $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3$ is 8 ppm/ $^\circ\text{C}$ which increases to 13.3 for $x = 0.05$ in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$. It is evident from the Fig. 4 that the values of τ_f for $x = 0.1$ and 0.15 in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ are 3.6 and -4.2 ppm/ $^\circ\text{C}$, respectively. This means that the temperature coefficient of resonant frequency is approaching zero between $x = 0.1$ and 0.15 in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$. In general it is observed that the τ_f values of the compositions becomes more and more negative with increasing concentration of Ti + W against Ta in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3$. The τ_f for $x = 1.0$ in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$, is -25.1 ppm/ $^\circ\text{C}$. This value is more than thrice the τ_f of $\text{Ba}(\text{Mg}_{1/3}\text{W}_{1/3}\text{Ti}_{1/3})\text{O}_3$ in a previous report [18] by Takahashi et al. (-7.6 ppm/ $^\circ\text{C}$), which is expected to be due to the presence of BaTiO_3 in the Ti + W rich compositions.

The unloaded quality factor of compositions in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ is plotted in Fig. 5. It is evident that the quality factor of the composition for $x = 1.0$ in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$, is inferior to that of the other end member, $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3$. The quality factor varies from $Q_u \times f = 100,500$ to 35,400 GHz as x varies from 0 to 1.0 in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$. The lower value of quality factor of the latter may be attributed to its relatively low-densification. It is worthwhile to note that for zero τ_f compositions in the present system ($0.1 < x < 0.15$ in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$), the quality factor is reasonably high ($Q_u \times f = 80,000$ – $90,000$ GHz).

4. Conclusions

The $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$ [$x = 0.01, 0.02, 0.03, 0.04, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9$ and 1.0] ceramics were prepared by the conventional mixed oxide route and their microwave dielectric properties were studied. The dielectric constant decreases, while the temperature coefficient of resonant frequency decreases and approaches negative values with increase of x in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$. The τ_f approaches zero between $x = 0.1$ and 0.15 in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$. The microwave dielectric properties of $\text{Ba}(\text{Mg}_{1/3}\text{W}_{1/3}\text{Ti}_{1/3})\text{O}_3$ are $\epsilon_r = 15.4$, $\tau_f = -25.1$ ppm/ $^\circ\text{C}$, $Q_u \times f = 35,400$ GHz. The zero τ_f compositions in the present investigation ($0.1 < x < 0.15$ in $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{(2-2x)/3}\text{W}_{x/3}\text{Ti}_{x/3})\text{O}_3$) have reasonably high quality factor ($Q_u \times f = 80,000$ – $90,000$ GHz) which could be used for practical dielectric resonator applications.

Acknowledgement

The authors acknowledge Defence Research Development Organization, New Delhi for financial support.

Reference

- [1] L. Chai, M.A. Akbas, P.K. Davies, in: Proceedings of the International Conference on Solid State Chemistry of Inorganic Materials, 2–3 December, Boston, 1996), pp. 443–449.
- [2] D.J. Barber, K.M. Moulding, J.I. Zhou, M. Li, J. Mater. Sci. 32 (1997) 1531–1544.
- [3] B.P. Burton, E. Cockayne, Phys. Rev. B 60 (1999) 12542–12546.
- [4] J. Browne, Microwaves RF 39 (2000) 153–158.
- [5] T. Nagai, T. Inuzuka, M. Sugiyama, Jpn. J. Appl. Phys. 31 (1992) 3132–3135.
- [6] J. Venkatesh, V.R.K. Murthy, Mater. Chem. Phys. 58 (1999) 276–279.
- [7] H. Kagata, J. Kato, Jpn. J. Appl. Phys. 33 (1994) 5463–5465.
- [8] J. Youn, K.S. Hong, H. Kim, J. Mater. Res. 12 (1997) 589–592.
- [9] H.J. Youn, K.S. Hong, H. Kim, J. Kor. Phys. Soc. 32 (1998) S524–S526.
- [10] W.K. Choo, S.B. Park, K. Eo, H.J. Im, in: Proceedings of the 10th US–Japan Seminar on Dielectric and Piezoelectric Ceramics, 2001, p. 79.
- [11] M. Furuya, A. Ochi, Jpn. J. Appl. Phys. 33 (1994) 5482–5487.
- [12] H. Takahashi, K. Ayusawa, N. Sakamoto, Jpn. J. Appl. Phys. 36 (1997) 5597–5599.
- [13] B.W. Hakki, P.D. Coleman, IRE Trans. Microw. Theory Tech. MTT-8 (1960) 402–410.

- 176 [14] W.E. Courtney, IEEE Trans. Microw. Theory Tech. MTT-18 (1970) 476–485.
- 177 [15] J. Krupka, K. Derzakowski, B. Riddle, J. Baker-Jarvis, Meas. Sci. Technol. 9 (1998) 1751–1756.
- 178 [16] S.J. Penn, N. McN Alford, EPSRC Final Report, EEIE, South Bank University, London, 2000.
- 179 [17] K.P. Surendran, P. Mohanan, M.T. Sebastian, (unpublished data).
- 180 [18] H. Takahashi, K. Ayusawa, N. Sakamoto, Jpn. J. Appl. Phys. 37 (1998) 908–911.
- 181 [19] E.J. Fresia, L. Katz, R. Ward, J. Am. Ceram. Soc. 81 (1959) 478–481.
- 182 [20] S.J. Penn, N.M. Alford, A. Templeton, X. Wang, M. Xu, M. Reece, K. Schrapel, J. Am. Ceram. Soc. 80 (1997) 1885–1888.