Microwave Dielectric Losses Based on Polarities

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Microwave dielectrics have been researched up to present, and many compounds with excellent properties are presented, which are listed in a book "Dielectric Material for Wireless Communication" written by Sebastian (2008). These materials are expected for usage of millimetrewave applications and new applications as paraelectrics, so origins of losses for microwave dielectrics are resumed based on the polars produced by impurities, defects, grain boundary, and stress originated from fluctuation of lattice constants. For new applications, the materials with low losses should be redesigned.

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I. INTRODUCTION

Microwave dielectrics including millimeterwave dielectrics have good properties of low dielectric losses that are high \( Q \), which are mainly belonging to paraelectrics. Sebastian (2008) presented a book named “Dielectric Materials for Wireless Communication” in which about 2300 species low loss dielectric materials and about 750 references are listed on the Appendix 2 in the book [1]. These many materials have been developed and applied for microwave applications such as resonators, filters, absorbers and LTCC. But, now the applications have been reducing though the properties are excellent for microwave communications. So, these materials should be applied for new applications [2–3].

In this paper, the most important property for microwave dielectrics that is low dielectric losses is discussed. Origins of loss will be produced by polarity, which are resumed.

II. MICROWAVE DIELECTRIC PROPERTIES

The microwave dielectrics are familiar with radio frequency wave. The low loss dielectric materials are resonated by microwaves in air and change it to electric current in circuit, and vice versa. It is well known that the microwave materials have three important characteristic properties such as high quality factor \( Q \), dielectric constant \( \varepsilon_r \) and temperature coefficient of resonant frequency \( TCf \). The most important property is low dielectric losses. The most of these materilas are paraelectrics without inversion symmetry \( i \), and have no

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defects and none impurities, which are perfect structure and fine compositions. Origin of losses will be discussed based on the polarities improved due to impurities, defects, grain boundary, and rattling effect as follows.

II-1. Impurities bring high losses

![Image](a) commercial forsterite with glassy phase. (b) Fine glass free forsterite wiht high Q.

In the case of forsterite, Tsunooka et al. [4] presented high $Q_f$ of 240000 GHz which was prepared using high purity low materials of 99.9 %. Commercial forsterite with $Q_f$ of 10000 GHz was including glass phase as shown in Fig. 1 (a). High $Q_f$ forsterite has clean grains without glass phases as shown in Fig. 1 (b). Impurities make dipoles, which produce dielectric losses by the inversion of the polar when microwave applied.

II-2. Defects bring high losses

Koga et al. [5–8] presented three area in the vicinity of pure $\text{Ba(Zn}_{1/3}\text{Ta}_{2/3})\text{O}_3$ (BZT): 1st one is ordered single phase, 2nd is ordered phase with secondary phase, and 3rd is disordered single phase with defects as shown in Fig. 2 [9]. The chemical formula of the disordered single phase in the 3rd area is presented, which include excess Ba ions and form defect of B-site and oxygen ions. Kugimiya [10] presented the chemical formula with defects as follows:

$$\text{Ba}_{1+\alpha} \left( \text{Mg}_{1/3}\text{Ta}_{2/3+\gamma} \text{V}_{\alpha-\gamma} \right) \text{O}_{3+\alpha+5\gamma/2}\text{V}_{2\alpha-5\gamma/2}.$$

Koga et al. [6] studied crystal structure and properties on the three lines in the small area within +/-3% in the vicinity of BZT as shown in Fig. 2. Now, the 3rd line S-R-Q-A-N-O-P line will use for the explanation in this paper. X-ray Powder Diffraction (XRPD) patterns are shown in Fig. 3 (a), and special diffraction peaks of 420 for cubic are shown in Fig. 3 (b). The patterns of S, R and Q as shown in Fig. 3 (a) were single phase of BZT with disordered
FIG. 2: Koga et al. presented three area in the vicinity of BZT. (I) Ordered single phase of BZT, (II) Ordered BZT with secondary phase, (III) Disordered single phase of BZT with defects. (1): $x$BaO·$(2-x)1/3$ZnO·$2/3$TaO$_5$, (2): BaO·$y1/3$ZnO·$(2-y)2/3$TaO$_5$, (3): $x$BaO·$1/3$ZnO·$(2-x)2/3$TaO$_5$, $x$ and $y$: composition deviation from BZT.

cubic crystal structure, and A, N, O and P have super lattice peaks as shown by asterisk "*" which show ordered trigonal crystal structure. Only A sample shows a single phase with super lattice, and other ones have additional diffraction peaks originated as secondary phase with asterisk "s". The phase changes are shown clearly in Fig. 3 (b) as presented by Kawashima et al. [11] Koga et al. also analysed Ordering ratios using Rietveld method as shown in Fig. 3 (c) [6–7].

Ordering ratio, density, dielectric constant and $Qf$ are shown in Fig. 4 (a), (b), (c) and (d) [6], respectively. Ordering ratio of cubic disordered phase in Ba-rich S, R and Q region were low of about 20% by Rietveld method as shown in Fig. 4 (a). The samples in this area show low sintering density with porosities accompanying low dielectric constants as shown in Fig. 4 (b) and (c). Porosities usually did not affect to the $Qf$ because of air with high $Qf$. Origin of low $Qf$ as shown in Fig. 4 (d) should be generated by defects in B-site and oxygen.

On the other hand, Kugimiya [10] presented Ba(Mg$_{2/3}$Ta$_{2/3}$O$_{15}$)$_{2/3}$V$_{1/3}$O$_3$ solid solutions without defects of A-site and oxygen on the tie line BMT-BaTa$_4/5$O$_{3}$. Vacancies in B-site are neutral without electrons. The highest $Qf$ composition is located on the near
FIG. 3: XRPD patterns (a) and characteristic diffraction peaks (b) of around 114 to 115°/2θ of compounds S to P as shown in Fig. 2. (c) Characteristic diffraction peaks analyzed by Rietveld method, the calculated and observed patterns are shown on the top by the solid line and the dots, respectively. The vertical marks below the patterns indicate the positions of disordered and ordered phases. The trace on the bottom is a plot or the difference between the calculated and the observed intensities.

position of the tie line as shown in Fig. 5, which is shifted from ideal BMT composition to BaTa$_{4/5}$O$_3$ direction. Origin of high $Qf$ will be due to high structural density as heavy atom Ta substituted for Mg. Koga et al. also stated the shift of the composition from A to E as shown in Fig. 2 [9].

II-3. Grain growth brings high $Qf$

Miyauchi et al. [12] presented a high $Qf$ value of Al$_2$O$_3$ by the grain growth. The value is improved from 370000 GHz by Alford et al. [13] to 680000 GHz as shown in Fig. 6 which is plotted as a function of grain size [14]. Fig. 7 (a) and (b) shows $Qf$ values and grain size, respectively, as a function of sintering temperature. The SEM photographs also
FIG. 4: Ordering ratio (a), density (b), dielectric constant (c), and $Q_f$ values (d) of composition S to P as shown in Fig. 2 as function of composition deviation $x$ from BMT.

presented in Fig. 8 (a), (b), (c), (d) and (e) sintered at 1200, 1300, 1400, 1500 and 1600 °C, respectively. The grain size and $Q_f$ values become large according to sintering temperature.

II-4. Compositional ordering also brings high $Q_f$

In the case of pseudo-tungstenbronze solid solutions [15–16], $Q_f$ as a function of composition $x$ on the Ba$_{6-3x}$R$_{8+3x}$Ti$_{18}$O$_{54}$ ($R$: rare earth) chemical formula shows highest value at $x = 2/3$ which is special point on the structure as shown in Fig. 9 (a) [17]. Structural formula of the solid solutions is [R$_{8+2x}$Ba$_{2-3x}$V$_x$]$_{A1}$[Ba]$_{A2}$Ti$_{18}$O$_{54}$. Here, $A1$ is rhombic site in perovskite block, and $A2$ is pentagonal site as shown in Fig. 10. When $x = 2/3$, Ba ions in perovskite block become zero, so compositional ordering occupying $R$ in perovskite block and Ba in pentagonal site is generated on this special point. In the region of $x < 2/3$, Ba ions are locating in perovskite block, and $x = 2/3$, only $R$ ions are locating in the perovskite block as shown in Fig. 10 (a) and (b), respectively.
These examples such as less impurities and less defects, grain growth, and compositional ordering bring high $Q_f$ values. Each example has less polarities as shown in Fig. 11. Impurity and defect produce polar with impurity and near ions as shown in Fig. 11 (a). The polar absorb energy under the microwave, so $Q_f$ was degraded. Usually microwave materials are made using high purity materials. Defects also degraded the $Q_f$ such as impurities, so sintering condition should be avoided reduction condition generated oxygen defects. The grain boundary also produce polar on the grain boundary as shown in Fig. 11 (b). Grain growth reduces grain boundary and single crystal has no grain boundary, so the $Q_f$ is become high. In the case of Al$_2$O$_3$, the single crystal sapphire show high $Q_f$ of 1170000 GHz parallel to $c$-axis and 1890000 GHz perpendicular to $c$-axis [18]. In the case of pseudo-tungstenbranze type compound, Ohsato et al. presented the highest $Q_f$ value at $x = 2/3$ (Fig. 9 (a)) which has less internal strain-as shown in Fig. 12 [15]. According to increasing of Ba-ion number in the perovskite block, the internal strain obtained from FWHM by X-ray powder diffraction increases. The internal strain is obtained from fluctuation of lattice parameter, that is, size of the unit cell including perovskite block with Ba becomes large. The expansion of the unit cell brings enlargement of polyhedra such as
FIG. 6: $Q_f$ values of $\text{Al}_2\text{O}_3$ as a function of mean grain size. Miyauchi improved the $Q_f$ value of $\text{Al}_2\text{O}_3$ from 370000 to 680000 GHz.

FIG. 7: $Q_f$ values (a) and mean grain size (b) of $\text{Al}_2\text{O}_3$ as a function of sintering temperature.

TiO$_6$ octahedra, which also promotes rattling effects of the polyhedra. So, the dielectric constants increase with increasing number of Ba-ions in perovskite block as shown in Fig. 9 (b). In this case also, the origin of loss is brought from enlarged polarites in perovskite...
FIG. 8: SEM photographs (a), (b), (c), (d) and (e) of sintered samples at 1200, 1300, 1400, 1500 and 1600 °C, respectively.

FIG. 9: $Qf$ values (a) and $\varepsilon_r$ (b) of pseudo-tungstenbronze type solid solutions as a function of $x$ on the $\text{Ba}_6-3xR_{8+2x}\text{Ti}_{18}\text{O}_{54}$ solid solutions.
III. CONCLUSIONS

Up to present, many microwave dielectrics about 2300 compounds have been developed as shown in “Dielectric Materials for Wireless Communication” written by Sebastian
[1]. Now, the microwave dielectrics expected for usage for millimeterwave region, and new applications. The usage for high frequency area needs more high $Q_f$, and new applications are expected based on the excellent properties, so the authors presented origin of low dielectric losses based on the many research. The origin of low losses comes from polarity produced by impurities, defects, grain boundary, and strain originated from fluctuation of lattice constants. When new applications appeared, the properties should be redesigned based on these origins.

FIG. 11: Dipole produced by impurity (a) and grain boundary (b).

FIG. 12: Strain of the pseudo-tungstenbronze as a function of composition $x$. 

Special composition
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