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Polaron conduction loss in microwave dielectric ceramics

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It has been generally accepted that the product of the unloaded quality factor and resonant frequency is the universal parameter for comparison of dielectric resonators with different size.[1,2] However, it is suggested in this study that this universal parameter should be modified due to the presence of the polarons. From the frequency dependence of the unloaded quality factor, it is possible to extract the factor determined only by the phonon scattering effects, and we denoted this parameter by Q_s. It was found that the Q_s parameter for $Zr_xSn_zTi_yO_4$ (ZST) and $Ba(Zn_{1/3}Ta_{2/3})O_3$ (BZT) ceramics showed constancy in the frequency range of 2–12 GHz, which supports the idea of polaron conduction loss contribution to the dielectric loss.

I. INTRODUCTION

The assessment of the microwave dielectric materials by the quality factor or dielectric loss is important to the engineers fabricating dielectric resonators. The origin of the dielectric losses of microwave ceramics is well described by Wersing[3] and can be generally categorized by three factors: anharmonic forces which interact with the crystal's phonon, periodicity defects which lead to quasibonded states, and dipole relaxation of impurities or relaxation of space charge polarization at interfaces. Before proceeding further, it would be worthwhile to refer to the third factor. There would be some dispute on the contribution of this factor because the dipole relaxation and space charge polarization mechanism would be frozen at the microwave frequency as pointed out by Kingery *et al.*[4] Therefore, it is suggested to ignore the third factor, and this is in accordance with many other researchers who consider only the phonon effect.[5,6] According to Wersing,[3] it is emphasized that the two former mechanisms can be seen as energy transfer from the exciting microwave and the wave vector of zero value to transverse optical phonons. This predicts the linear increase of losses with frequency which is a characteristic for phonon effects. However, the donor type periodicity defects (e.g., oxygen vacancy) ionize to give electrons to the conduction band. This can be envisioned by the band model with dopant level near the conduction band. The electrons in ionic compounds interact with the polar modes of the crystals to form polaron.[7] This particle acts as a charge carrier, so it may be possible for polaron to contribute to conduction loss in microwave frequency. It is well known that there are two kinds of polarons: large polaron and small polaron. Because small polaron behaves much like ion in the aspect of relatively small mobility at room temperature, it can be induced that, in the microwave frequency, this mechanism would be frozen like ionic conduction mechanism. Therefore, consideration of the large polaron conduction would be enough for microwave frequency. As suggested by Hench and West,[8] the displacement occurred by polaron formation creates an energy level just below the conduction band ($\sim$0.1 eV less than conduction band level). The above argument would be exactly the same for acceptor-type defects except that the mobility of polarons in this case is lower than the above case. Therefore, the movement of the polarons will be like electrons or holes in extrinsic semiconductor. This suggests that this factor may take a portion of dielectric loss for extremely low loss materials used in microwave resonators. In this study, the contribution of polaron conduction to the dielectric loss will be discussed using a semiquantitative approach, and the validity of this argument is performed by comparing the published data with the loss equation modified for polaron conduction.

II. EXPERIMENTAL

Systematic experimental data are rarely available on the frequency dependence of the dielectric loss. However, Wersing[3] has published a series of such data. Figure 1 shows the product of quality factor and frequency as a function of frequency for $Ba(Zn_{1/3}Ta_{2/3})O_3$ (BZT), $Zr_xSn_zTi_yO_4$ (ZST), and Nd_2O_3–TiO_2–BaO–Bi_2O_3 (NBT) resonators. It is reported that the unloaded quality factor measured by resonant cavity with appropriate dimension becomes essentially equal to the quality factor originated solely from the dielectric material itself,

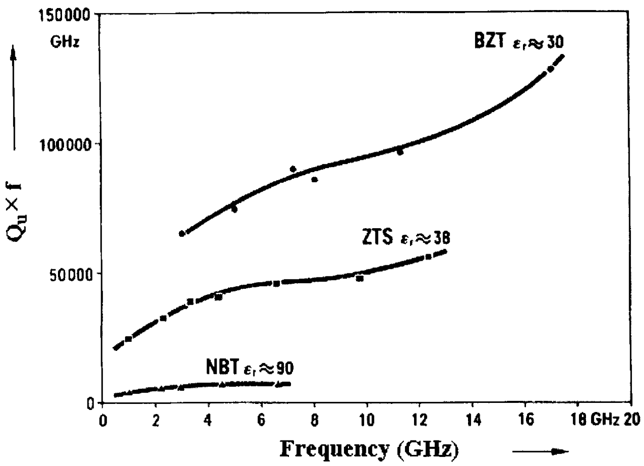


FIG. 1. Product of quality factor and frequency as a function of frequency for the resonators from the three material systems: $Ba(Zn_{1/3}Ta_{2/3})O_3$ (BZT), $Zr_xSn_zTi_yO_4$ (ZST), and $Nd_2O_3-TiO_2-BaO-Bi_2O_3$ (NBT) (see Ref. 3).

which is in turn the reciprocal of the loss tangent.[9] Therefore, if the phonon effect is the dominant one responsible for the loss, the measured product of quality factor and frequency should remain constant with respect to frequency. However, as shown in Fig. 1, this product increased with resonant frequency. The author mentioned this phenomenon and explained that some relaxation losses still play a role in these ceramics. Within the context of the author's paper, it is reasonable that the slight increase of the product is attributed to the dipole relaxation or the space charge polarization.[3] However, in our opinion, this is not likely the case since it is usually doubtful whether these mechanisms will play a role in the losses at the frequency up to $\sim 10^9$ Hz as mentioned by Kingery *et al.*[4] There should be a more reasonable mechanism to explain the deviation of the product of quality factor and frequency from the constancy.

III. RESULTS AND DISCUSSION

Therefore, it is proposed that polaron conduction is also responsible for the loss in low loss dielectric materials applied for microwave resonators. Most conventionally sintered ceramics contain defects which produce defect levels inside the band gap. These defects are easily ionized to give charge carriers: electrons or holes. In polar crystals, such an ionized defect polarizes the lattice around it by local distortion. This polarization creates a local electric field which self-localizes the charge by lowering its energy.[10] The electron and local deformation field make up the polaron, a concept reviewed by Bunget and Popescu.[11] If the polarization zone extends over a region of the order of the lattice constant, there exists a low radius polaron (small polaron), and if it extends over several lattice constants, the ensemble is a high radius polaron (large polaron). It is expected that only the large polaron will respond to the time-varying electromagnetic field in microwave frequency. Therefore, we will discuss the phonon effect accompanied by the large polaron conduction on the dielectric loss in microwave frequency.

The quality factor of the dielectric resonator originated from several sources can be expressed as follows:

$$\frac{1}{Q_u} = \frac{1}{Q_1} + \frac{1}{Q_2}\,, \qquad (1)$$

where Q_u is the unloaded quality factor, Q_1 is the quality factor originated from the polaron conduction, and Q_2 is the quality factor originated from the phonon scattering effects. It is well known that the frequency dependence of the conduction loss and the phonon scattering loss is different in the following manner.[3,4]

$$Q_1 = C_1 \times f\,, \qquad Q_2 = C_2 \times \frac{1}{f}\,, \qquad (2)$$

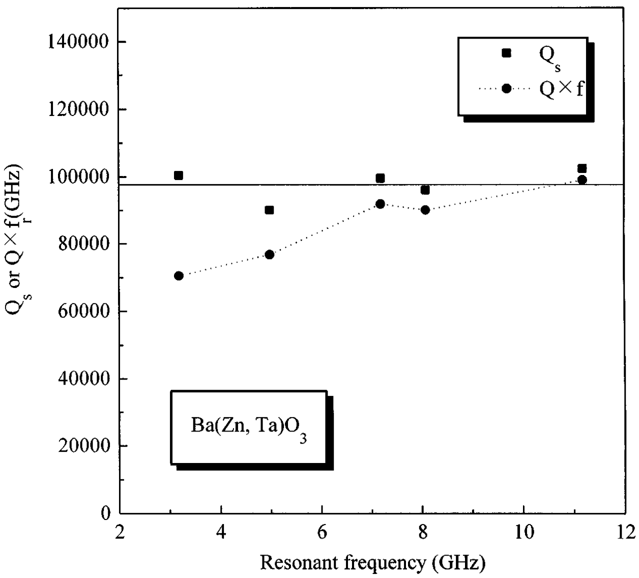
where C_1, C_2 are constants and f is frequency.

The presence of the polaron conduction lowers the effective unloaded quality factor, Q_u, originated solely from the dielectric resonator. This factor is introduced by external factor such as ionized defects coming from the sintering process. Therefore, it is worth defining a parameter which only concern the internal factor, and sets the maximum limit for the unloaded quality factor. We will denote the constant C_2 in Eq. (2) by Q_s which is often referred to as the product of quality factor and frequency. This factor can be rearranged with the help of Eqs. (1) and (2).

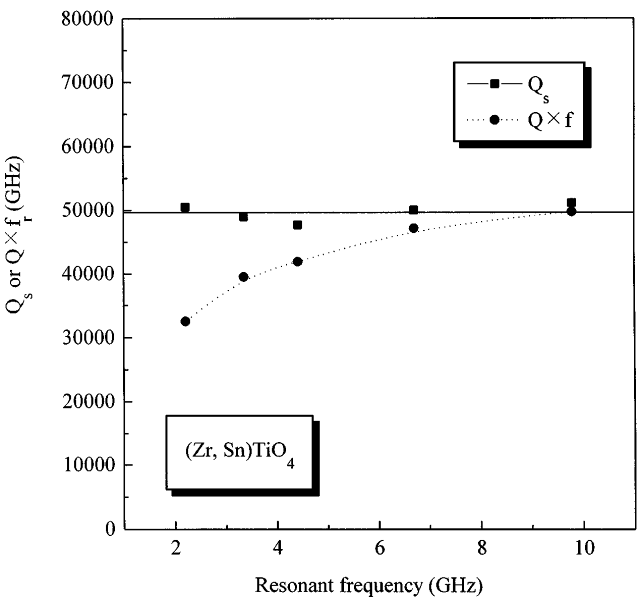
$$Q_s \equiv C_2 = Q_u \times f + \frac{C_2}{C_1}\frac{Q_u}{f}\,. \qquad (3)$$

The physical meaning of this parameter is that it is originated only from the ion vibration and deformation which causes an energy transfer to thermal phonons. From the plot of the $Q_u \times f$ vs Q_u/f, the two constants C_1 and C_2 can be determined from linear fitting using MICROCAL ORIGIN software.

Figure 2 shows the variation of the Q_s and the $Q_u \times f$ with the frequency. NBT resonator was not considered, because the variation of the data point with frequency was comparable to the dimension of the data point symbol itself. The values of C_1 and C_2 were 22,680 and 97,686, respectively, for BZT and 16,088 and 49,680, respectively, for ZST. As can be seen from the plot, the constancy of Q_s factor is well observed for ZST and BZT materials in the frequency range of 2–12 GHz. This supports the validity of Eq. (3). Also, it is seen that the $Q_u \times f$ factor approaches Q_s value as the frequency increases. This is well explained by the



(a)



(b)

FIG. 2. Comparison of the Q_s factor and the $Q_u \times f$ factor dependence on the resonance frequency for (a) BZT and (b) ZST.

fact that the contribution of Q_1 to Q_u becomes negligible as the frequency increases.

The presence of the polarons from defects restricts us from using the $Q_u \times f$ factor as a universal parameter in comparing different sizes of dielectric resonators. Instead, the Q_s parameter suggested in this study is its modified form without adding or losing its physical meaning. Also, it shows the limit of the quality factor for given ceramic dielectric resonators made by conventional sintering process.

IV. CONCLUSIONS

The contribution of polaron conduction to the dielectric loss was discussed. From the frequency dependence of the unloaded quality factor, it was speculated that the polaron conduction will take place in addition to the phonon scattering process. Moreover, Q_s parameter was introduced which defines the product of resonant frequency and quality factor originated only from the ionic dipole vibration and deformation. This parameter can be used not only as a universal parameter for comparison of dielectric resonators with different size, but also as a processing limit of the unloaded quality factor for conventionally sintered dielectric resonators.

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