Ultralow loss polycrystalline alumina

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Polycrystalline alumina with extremely low microwave dielectric loss is reported with properties analogous to a theoretical ensemble of randomly oriented, single crystal sapphire grains. By avoiding deleterious impurities and by careful control of microstructure, we show that grain boundaries in aluminum oxide have only a limited influence on the dielectric loss. A method of measuring the electric permittivity and loss tangent of low-loss microwave ceramic dielectrics is reported. The electrical parameters such as relative permittivity and loss tangent are extracted using the radial mode matching technique. The measured values for ultralow loss polycrystalline aluminum oxide agree well with theoretical values modelled on an ensemble of randomly oriented anisotropic single crystal sapphire grains. © *2002 American Institute of Physics.*
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Microwave dielectric ceramics with extremely low loss ($\tan \delta < 10^{-5}$) and relatively low permittivity ($\varepsilon_r \sim 10$) such as sapphire (Al_2O_3) and MgO, have applications in microwave communication in the 10–50 GHz frequency band. They also have application as resonators for ultrastable oscillators in clocks. Single crystals are used due to their low losses in the range of 10^{-5} at 10 GHz.[1] Polycrystalline analogs of these materials usually have similar permittivity, but losses that are generally a factor of at least two higher. The measurement of low loss, low permittivity microwave dielectric ceramics using the $TE_{01\delta}$ is limited in accuracy by the electric filling factor and geometric factor of the conducting shield. By carefully choosing the dimensions of the conducting shield for optimum electric filling factor P_e and geometric factor G, it is possible to measure the $\tan \delta$ with 10% accuracy for $\tan \delta > 10^{-5}$.[2] The preferred method of measurement for $\tan \delta < 10^{-6}$ utilizes high order whispering gallery modes,[3,4] which have electric filling factors approaching unity and very high geometric factors. Unfortunately, the modal number density is usually very high in the frequency range in which whispering gallery modes exist, and careful choice of cavity dimensions is necessary to avoid spurious modes.[5] This letter seeks to extend the usefulness of the $TE_{01\delta}$ mode measurement method by eliminating a great source of error: the support. The relative permittivity of the dielectric resonator can be calculated using the radial mode matching method.[6] Instead of solving for frequency, as is usually done, one can instead solve for ε_r. After calculation of the electric filling factor and geometric factors, the $\tan \delta$ can be obtained from the expression

$$\tan \delta = P_e^{-1}(Q_0^{-1} - R_s G^{-1} - P_{\text{spacer}} \tan \delta_{\text{spacer}}), \quad (1)$$

where P_e is the electric filling factor, Q_0 is the unloaded quality factor, R_s is the surface resistivity, and G is the geometric factor.

An alumina dielectric resonator was constructed as shown in Fig. 1 using the TiO_2-doped Al_2O_3 developed in the laboratory.[7] This material is high purity and fine grained, both of which are necessary in order to achieve very low loss.[8,9] The dimensions of the conducting shield are diameter 36.00 mm, height 23.48 mm. The alumina puck has diameter 10.69 mm and height 4.34 mm. The alumina spacer has diameter 4.13 mm and height 6.77 mm.

The resonant frequency of the quasi-TE_{011} ($TE_{01\delta}$) mode was measured at room temperature (300 K) and found to be 8.95 557 GHz. The measurement was performed in transmission (S_{21}) using an Agilent network analyzer (HP8722). Input and output coupling to the resonator was achieved using coaxial transmission lines with small loops formed by soldering the central conductors to the outer shield of the coaxial cable. The coupling was -40.5 dB, measured from the insertion loss. The loaded quality factor Q_L of the mode was measured and found to be 62 500. For equal input and output couplings, the unloaded quality factor Q_0 can be calculated as

$$Q_0 = \frac{Q_L}{1 - |S_{21}|}. \quad (2)$$

In this case, Q_0 was found to be 63 090. The resistivity of the oxygen-free copper shield was found by measuring Q_0 for the TE_{111} and TE_{011} modes of the empty cavity (without dielectric). This was done in order to confirm the frequency dependence. These measurements were performed as a function of temperature using a Gifford–McMahon closed-cycle cooler from 20 to 320 K. At room temperature and 8.95556 GHz, the resistivity was calculated to be 2.24×10^{-8} Ω m. The surface resistance R_s in this case was 28.15 mΩ.
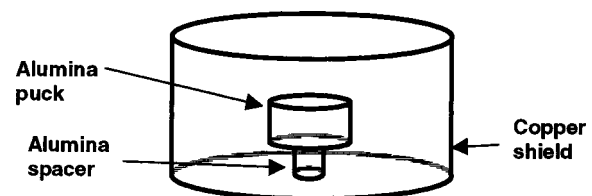

FIG. 1. Structure of alumina dielectric resonator.

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TABLE I. Comparison of filling factors and geometric factors using different modelling methods.

	Mode Matching	MAFIA	Finite difference time domain
Electric filling factor P_e	0.777	0.770	0.775
Geometric factor G	2812	2669	2825

The ε_r of the alumina was calculated using the radial mode matching method. This entails solving a linear system of equations for a prescribed number of eigenmodes N. For this problem, $N=9$ modes were found to be sufficient for convergence of the solution. The value of ε_r in this case for polycrystalline alumina is 10.15. The $\tan\delta$ can be calculated using the expression

$$\tan\delta = P_e^{-1}(Q_0^{-1} - R_s G^{-1}). \tag{3}$$

After measuring the unloaded quality factor Q_0 and surface resistivity R_s, it is necessary to calculate the electric filling factor and the geometric factors, using

$$P_e = \frac{\iiint\limits_{V_d} \varepsilon_r \mathbf{E}\cdot\mathbf{E}^* dV}{\iiint\limits_{V} \varepsilon_r(V)\mathbf{E}\cdot\mathbf{E}^* dV}, \tag{4}$$

and

$$G = \frac{\omega\iiint\limits_{V} \mu_0 \mathbf{H}\cdot\mathbf{H}^* dV}{\oiint\limits_{S} \mathbf{H}_t \cdot \mathbf{H}_t^* dS}. \tag{5}$$

These quantities can be evaluated once the field coefficients of the series expansion are known. Surface and volume integration of the fields allows one to calculate P_e and G. Alternatively, the incremental frequency rule can be used.[9,10] The values of P_e and G were calculated to be 0.777 and 2812, respectively. These values were compared with results from a two-dimensional finite difference time domain code[11] and MAFIA.[12] These results are given in Table I. From G and R_s we derive a conductor quality factor Q_c of 99 893. This results in a dielectric quality factor Q_d of 171 241. We can now calculate the loss tangent,

$$\tan\delta = \frac{1}{P_e Q_d}. \tag{6}$$

The result is $\tan\delta = 7.516\times10^{-6}$ at a frequency of 8.955 56 GHz. Assuming a linear frequency dependence for the loss in the frequency range from 8 to 10 GHz, allows the loss tangent to be normalized to 10 GHz (8.393×10^{-6}). Hence a Qf product of 1 191 580. The properties are summarized in Table II.

For comparison, Krupka et al.[3] reported a relative permittivity tangential to the c-axis, ε_t, of 9.400, and parallel to the axis, ε_z, of 11.587 for single crystal sapphire. Also reported[4] were values for the $\tan\delta$ for the transverse plane,

TABLE II. Dielectric properties of polycrystalline TiO$_2$-doped Al$_2$O$_3$.

Relative permittivity ε_r	10.15
$\tan\delta$ measured at 8.95557 GHz	7.516×10^{-6}
$\tan\delta$ normalized to 10 GHz	8.393×10^{-6}

$\tan\delta_t$ of 9.09×10^{-6} at 9.02985 GHz, and 4.76×10^{-6} at 8.85522 GHz for the axial component $\tan\delta_z$. By assuming that the frequency dependence of the loss tangent is linear, we can normalize all these values to 10 GHz for comparison. The normalized $\tan\delta$ for the results reported by Krupka et al.[3] are transverse $\tan\delta = 1.01\times10^{-5}$ and axial $\tan\delta = 5.38\times10^{-6}$. An effective loss tangent can be calculated in the same way as the permittivity; using the equation for effective permittivity, the result is $\tan\delta = 8.33\times10^{-6}$. It is interesting to note that the $\tan\delta$ value for the polycrystalline alumina measured here and normalized to 10 GHz is $\tan\delta = 8.39\times10^{-6}$.

The problem of the effective permittivity of an ensemble of randomly oriented uniaxial spherical crystallites has been investigated by numerous authors. The following equation was derived by Herring,[13]

$$\varepsilon' = \frac{1}{3}\varepsilon_z + \frac{2}{3}\varepsilon_t - \frac{2}{9}\frac{(\varepsilon_z-\varepsilon_t)^2}{\varepsilon_z+2\varepsilon_t}, \tag{7}$$

where the strong isotropy condition is assumed. A generalized form for the biaxial case was reported by Stroud:[14]

$$\varepsilon' = \bar{\varepsilon} - \frac{1}{9\bar{\varepsilon}}\sum_{i=1}^{N}(\varepsilon_i - \bar{\varepsilon})^2, \tag{8}$$

which is identical in form to that reported by Landau et al.[15] for a dielectric mixture.

Using the sapphire measurements of Krupka et al. to calculate the properties of polycrystalline alumina yields a relative permittivity ε_r of 10.094 and loss tangent $\tan\delta = 8.33\times10^{-6}$. We show that high purity, fine grained polycrystalline alumina possesses very low dielectric loss. A $\tan\delta$ of 8.33×10^{-6} was measured at 10 GHz and at 300 K. Using literature values for the $\tan\delta$ of sapphire, parallel and normal to the c-axis, we modelled the dielectric properties. We obtained very good agreement between the results of the model and the experimental observations.

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