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Tayloring microstructure for microwave dielectric properties - theory and experiment

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Pressureless sintering of cold isostatically pressed ceramic compacts into bodies of almost theoretical density is one of the most convenient methods to produce low loss dielectrics. The evolution of the dielectric properties is accompanied by simultaneous transformation processes in the microstructure. The interrelationship between these processes is discussed by introducing two independent factors. First the configurational factor modifies the dielectric properties by altering the relative composition of different phases (including pores) in material. Second, the intrinsic factor acts upon the effective dielectric properties by changes in the dielectric properties of the individual phases. Equations to describe the configurational factor are specified for limiting cases most relevant to sintered ceramics. Experimental data from microwave measurements at 30-40 GHz is analyzed for Magnesium-Aluminium-Spinels. It shows that the dielectric permittivity is fully controlled by the configurational factor. The intrinsic factor, however, gives a major contribution to the dielectric loss value.

1. INTRODUCTION

Ceramic materials are being intensively characterized and optimized with respect to their linear dielectric properties in the microwave and millimeter (mm-) wave range, because a steadily growing spectrum of applications relies on a tight control of these properties. Ceramics with low microwave and mm-wave absorption are used as substrates for integrated circuits [1], as substrates for high Tc superconductor films [2], and as electromagnetic windows [3].

The two basic parameters concerned are the permittivity (ε') and the dielectric loss tangent ($\tan\,\delta$). Whereas the permittivity determines the general signal transmission characteristics (signal speed, cross talk, reflection at interfaces), the dielectric loss tangent governs the signal attenuation. In practice, the control of the loss tangent is more critical since it can vary for the same material by an order of magnitude.

Previous work [4] has indicated that the succesful control of the dielectric properties necessarily involves the assessment of the development of the microstructure. In this respect, the technologically simpler method of cold pressing and sintering - when it leads to dense bodies - provides a wide spectrum of processing parameters and related microstructures. This work is an attempt to provide a general framework based on two essential aspects that can be used as a guideline to evaluate the influence of microstructure on the dielectric properties.

2. THE CONFIGURATIONAL FACTOR

Ceramics prepared by powder technological methods are generally two- or multiphase materials because pores have also to be considered as an individual phase. Assuming at this stage that the properties of the individual phases are not affected by the fabrication process, then the effective dielectric properties of the composite are only influenced by the content and orientation of the phases. This part of the microstructural tayloring will be defined here as the configurational factor.

A rigorous analysis is however hardly manageable as it would involve a complete stereological description of the phases (concentration, shape and orientation) and the solution of the adequate mathematical model. A valid approach for practical use is therefore to assume spherical particles or pores and to calculate the effective property of the multiphase material by applying iteratively the two phase formulations.

In this case it can be shown for low loss dielectrics that the major existing models can be approximated by the following relationship [5].

Case I Two phases with similar permittivity and arbitrary concentration c_1, c_2

$$(\varepsilon_2' = \varepsilon_1' (1 - \Delta)\ with\ |\Delta| \ll 1)$$

$$\varepsilon' = \varepsilon_1' \left(1 - c_2 \Delta (1 + a)\right), where\ a = \frac{\Delta}{3} (1 - c_2) \quad (2.1)$$

$$\tan\delta = \left(1 + \frac{\Delta}{3} c_2\right) c_1 \tan\delta_1 + \left(1 - \frac{\Delta}{3} c_1\right) c_2 \tan\delta_2 \quad (2.2)$$

$$\approx c_1 \tan\delta_1 + c_2 \tan\delta_2 \quad (2.3)$$

Case II Small concentrations of a second phase with arbitrary permittivity in a matrix formed by the first phase ($c_2 \ll c_1$)

$$\varepsilon' = \varepsilon_1' + \frac{3\varepsilon_1'}{\varepsilon_2' + 2\varepsilon_1'} (\varepsilon_2' - \varepsilon_1') c_2 \quad (2.4)$$

$$\tan\delta = (1 - 3c_2 A) \tan\delta_1 + 3c_2 A \tan\delta_2 \quad (2.5)$$

$$with\ A = 3(1 - \Delta)/(3 - \Delta)^2$$

The special case of a porous material is resolved by setting $\varepsilon'_2 = 1$; $\tan\delta_2 = 0$ and $c_2 = P$ (fractional porosity) in equations 2.4 and 2.5.

Experimental work with AlN [6], Mg-Al spinels [7], BN and Al_2O_3 [8] has shown that the measured values of permittivity can be well predicted by considering the configurational factor only (Fig. 1).

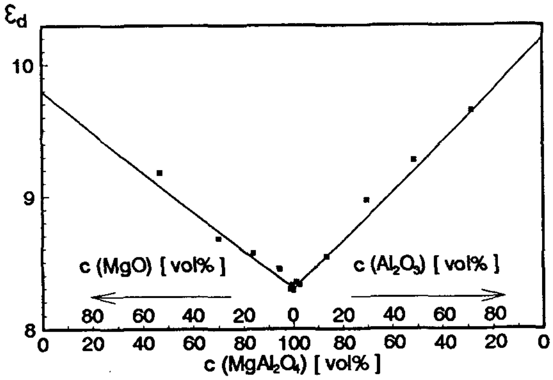


Figure1. Influence of stoichiometry on the permittivity of Spinel at 30 GHz extrapolated to theoretical density.

With regard to the dielectric loss, it has been found that the configurational factor is inadequate to understand the loss behaviour of the material (with the exception of BN - see Fig. 2 - which does not densify and undergo microstructural changes during conventional sintering). This is most obvious in the case of those materials where porosity is the only additional phase; as tan δ should vary by only 10% for P < 10% (see equation 2.5). In fact, significant changes are observed at comparable levels of porosity in AlN and Mg- Al spinel [4].

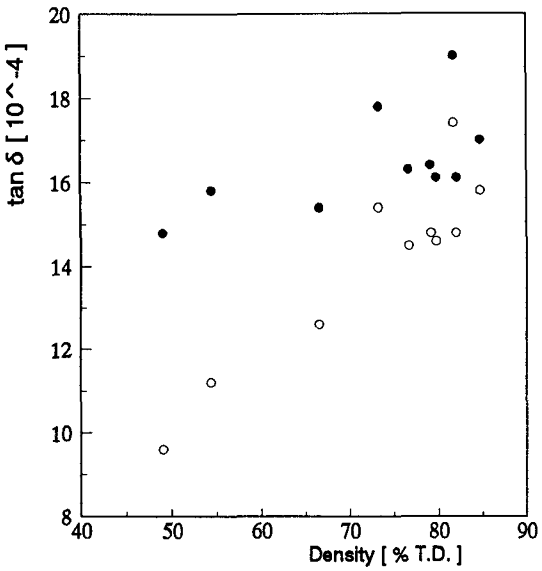


Figure 2. Dielectric loss at 30 GHz in pressed and sintered BN.
∘ measured
• extrapolated to theoretical density using equation 2.2

3. THE INTRINSIC FACTOR

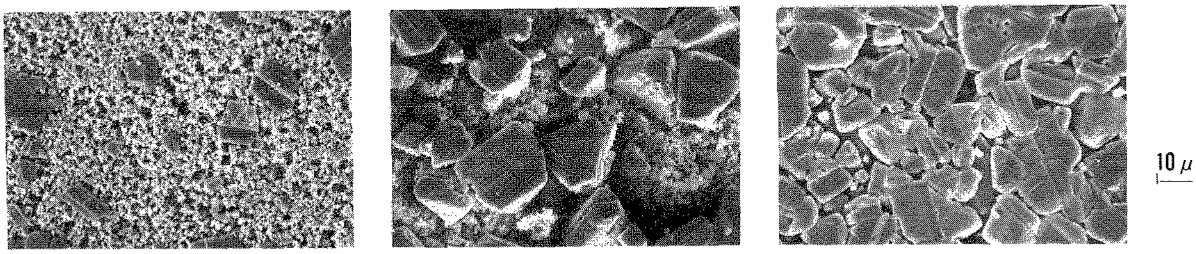
The fabrication process and parameters have in a general a complementary influence on the effective dielectric properties as they can alter directly the properties of the constituent phases. This part of the microstructural tayloring will be defined here as the intrinsic factor. In a single phase material this factor provides the only control on the dielectric properties.

Basically the dielectric properties are determined by the interaction of electromagnetic fields with the microscopic structure of the ceramic. In principle free charge carriers, localized bound charges (dipoles), collective lattice vibrations (phonons) and the electronic structure of the ions can be excited. In so far as the fabrication process reduces the strength of the interaction or number of interacting particles, it affects the intrinsic factor.

For the case of low loss dielectrics in the microwave range, which is intermediate between dipole relaxation and phonon resonance frequencies, only the tail of the resonant structure is operative. As a consequence, permittivity is hardly influenced by microscopic features, whereas the dielectric losses are still sensitive. Information on the intrinsic factor can be obtained by varying the sintering conditions. At least two effects are observed in the microstructure, restructuring of the grain morphology and grain growth (see fig. 3). In all materials investigated thus far, grain restructuring was found to be an essential precondition to reduce losses. Small additional improvements are achieved by extended thermal treatment which go along with grain growth. The observations can be rationalized with the concept that the intrinsic factor is closely related to the degree of perfection of the grains. Apparently the powder particles have a large number of lattice defects whose healing is very effective during the formation of new grains and proceeds at a lower rate once the grains are formed. The negative effect of lattice defects on loss is intuitively clear as they may act as dissipative dipoles and interaction partners for phonons.

4. SUMMARY AND CONCLUSIONS

The role of the fabrication process on the microwave dielectric properties can be distinguished between a configurational and intrinsic factor. The configurational factor can be quantified once the phase composition and properties are known. It is the major factor for tayloring the permittivity. The intrinsic factor which is most relevant for dielectric loss is most sensitive to perfection of the grains and so probably to lattice defects.



1630°C/5 h (tan $\delta = 3.4 \pm 0.1 \cdot 10^{-4}$)   1630°C/16.7 h (tan $\delta = 1.3 \pm 0.1 \cdot 10^{-4}$)   1630°C/20 h (tan $\delta = 1.1 \pm 0.1 \cdot 10^{-4}$)

Figure 3. Reduction of dielectric loss in sintered Mg-Al Spinel due to grain restructuring (intrinsic factor)

5. ACKNOWLEDGEMENT

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