MICROWAVE MEASUREMENTS OF DIELECTRIC CONSTANTS FROM COMPOSITE SAMPLES

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Abstract

Microwave dielectric constants of ceramic dispersions in the polyethylene matrix are studied by five well known mixture equations in this article. Ceramic powder of calcium titanate (CaTiO₃) filler with dielectric constant of 170 at 10 GHz is investigated. The experimental dielectric constant values of the mixtures are compared to the theoretical curves of the six mixing rules with basic particle shapes. The dielectric constant of pure ceramic is then estimated by these mixing laws from the dielectric constants of composites of various filler concentrations. The theoretical errors of estimation of various rules are derived and analyzed. The most adequate mixture law for measuring the dielectric constants of ceramic fillers will be recommended.

1 Introduction

This article reports on a study of the dielectric constant measurements of high dielectric constant ceramic at microwave frequency by powder mixture equations. The dielectric constant of calcium titanate ($CaTiO_3$) ceramic is indirectly obtained from the dielectric constants of ceramic-polymer composites. Dielectric properties of heterogeneous mixtures have been investigated by different authors for over one century [1-8]. Theoretically, the dielectric constant of a pure ceramic can be calculated from that of the ceramic-polymer composite if the mixing rule has a good accuracy. One can estimate the dielectric constant of a pure ceramic by simply measuring a composite sample which saves the high temperature sintering procedure of making a pure ceramic sample for measurements.

Recently, a new theoretical analysis method on the accuracy of calculating the dielectric constant from the composite samples by different mixture equations was studied for three basic particle shapes of fillers in reference [1]. The studied dielectric constants in that research are less than 40. Polyethylene powder was chosen as the matrix material. The dielectric constants at microwave frequency of the ceramic-polyethylene composite samples with various compositions were measured at 10 GHz. The dielectric constants of composites are then converted to the dielectric constants of pure filler ceramic medium by these mixing laws. However, there is no study on the materials with high dielectric constants using the mixture rules. The high dielectric constant powder of calcium titanate is then chosen as the study target in this paper. The experimental results will be applied to these equations to measure the

microwave dielectric constants of ceramics in this research. The new theoretical analysis of calculation accuracy will also be discussed.

The calcium titanate ceramic filler has a dielectric constant 170 at 10 GHz. Experiment results are compared with the theoretical curve of the six mixing laws of basic particle shapes. Furthermore, the dielectric constants of composites are then converted to the dielectric constant of pure filler medium by these mixing laws. The accuracy on estimating microwave dielectric constant of pure ceramic by various mixture rules will be compared. The most adequate mixture rule to calculate the dielectric constant of pure ceramic powder will be suggested.

2 Mixture equations

For composite materials containing dispersed spheres with low filler concentration, the mixture rule can be expressed as [2-5],

$$\frac{\varepsilon_{mix}}{\varepsilon_m} = \frac{\varepsilon_d(1+2V_d)+2\varepsilon_m(1-V_d)}{\varepsilon_d(1-V_d)+\varepsilon_m(2+V_d)} \sim \text{rule 1, spheres with low } V_d \tag{1}$$

where ε_{mix}, ε_d, and ε_m are dielectric constants of a composite, pure dielectric filler, and polymer matrix material, respectively. The V_d is the volume percentage of the filler medium. For high filler concentration [2-5],

$$\frac{\varepsilon_{mix}-\varepsilon_m}{3\varepsilon_{mix}} = \frac{\varepsilon_d-\varepsilon_m}{\varepsilon_d+2\varepsilon_{mix}}V_d \sim \text{rule 2, spheres with high } V_d \tag{2}$$

For filler particles with long prolate spheroids, such as cylinders, needles, or rods, random distribution of filler particles leads to the following equation [4,5],

$$\varepsilon_{mix}-\varepsilon_m = \frac{(\varepsilon_d-\varepsilon_m)(\varepsilon_d+5\varepsilon_m)}{3(\varepsilon_d+\varepsilon_m)}V_d \sim \text{rule 3, prolate spheroids with low } V_d \tag{3}$$

and,

$$1-V_d = \frac{(\varepsilon_d-\varepsilon_{mix})(2\varepsilon_m+\varepsilon_d+3\varepsilon_{mix})}{(\varepsilon_d-\varepsilon_m)(\varepsilon_d+5\varepsilon_{mix})} \sim \text{rule 4, prolate spheroids with high } V_d \tag{4}$$

The mixture rule for filler with the shapes of extended oblate spheroids, such as lamellae, flakes, and disks, can be expressed as [4-6],

$$\varepsilon_{mix}-\varepsilon_m = \frac{(\varepsilon_d-\varepsilon_m)(\varepsilon_m+2\varepsilon_d)}{3\varepsilon_d}V_d \sim \text{rule 5, oblate spheroids with low } V_d \tag{5}$$

and,

$$\frac{\varepsilon_{mix}}{\varepsilon_d} = \frac{3\varepsilon_m+2V_d(\varepsilon_d-\varepsilon_m)}{3\varepsilon_d-V_d(\varepsilon_d-\varepsilon_m)} \sim \text{rule 6, oblate spheroids with high } V_d \tag{6}$$

Fig. 1 shows the variations of the computation errors ($\Delta\varepsilon_d / \varepsilon_d$) with respect to the volume percentage of dispersed material assuming a 10% error of mixture rule ($\Delta\varepsilon_{mix} / \varepsilon_{mix}$) and $\varepsilon_d = 100$. It can be seen that the error increases with decreasing the volume concentration of dispersed material. Therefore, to have a more accurate estimation, higher volume percentage of V_d is a better choice. Obviously, mixture rules 1 and 2 are not good choice for calculating the dielectric constants of pure ceramics unless the rule is very accurate ($\Delta\varepsilon_{mix} / \varepsilon_{mix}$ very small). No obvious difference on the theoretical error was observed among rules 3 to 6. The accuracy will depend on the physical value of $\Delta\varepsilon_{mix} / \varepsilon_{mix}$ for individual mixture equations.
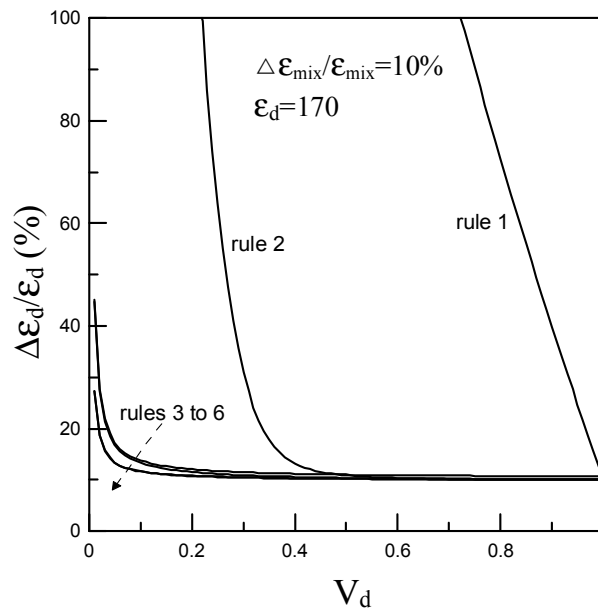

Figure 1. Error of calculating dielectric constant of pure ceramic.

3 Measurements

Suitable amounts with the desired volume concentrations of polyethylene and ceramic powders (V_d) were mixed using alcohol as a solvent and ZrO_2 balls in a plastic jar by ball milling more than 10 hours. After the well mixed suspension was stirred and evaporated until almost dry, it was baked at 80°C for 4 to 5 hours to obtain the complete dried powder mixture. The mixed powder was then pressed using a steel die at temperature 125°C (the melting point of polyethylene is 115°C) and under a pressure ~7,000 lb/in² for about 0.5 hour to make disk composite samples for measurements. The concentrations were calculated using the densities of polyethylene (0.915g /cm3) and ceramics, and the weight content of ceramics within the composite samples.

For the measurement of microwave dielectric constants, the post resonance method is adopted. Originally suggested by Hakki and Coleman in 1960 [9], this method has been widely used and has become the most popular dielectric resonance method for measuring the dielectric properties of dielectric samples. A cylindrical dielectric rod is placed between two parallel metal plates as shown in Fig. 2. Two coupling antennas are used to couple the power in and out. The S_{21} parameter is measured. The TE_{011} mode is adopted for measurements. The resonant frequency f_o, the half power bandwidth Δf_{3dB}, the insertion loss S_{21}, and the diameter $D(= 2a)$ and thickness L of the specimen are recorded for the calculations of

dielectric properties. The dielectric constant is calculated by the following equations of TE$_{011}$ mode [10],

$$\varepsilon_r = (\frac{\lambda_o}{2\pi})^2 (k_{ci}^2 + k_{co}^2) + 1 \tag{7}$$

$$k_{co}^2 = (\frac{2\pi}{\lambda_o})^2 [(\frac{\lambda_o}{2L})^2 - 1] \tag{8}$$

$$\frac{J_0(k_{ci}a)}{J_1(k_{ci}a)} = -\frac{k_{co}a}{k_{ci}a} \frac{K_0(k_{co}a)}{K_1(k_{co}a)} \tag{9}$$

$$\lambda_o = \frac{c}{f_o} \tag{10}$$

where k_{ci} and k_{co} are sample geometry dependents, J and K are Bessel function and modified Bessel function, respectively.
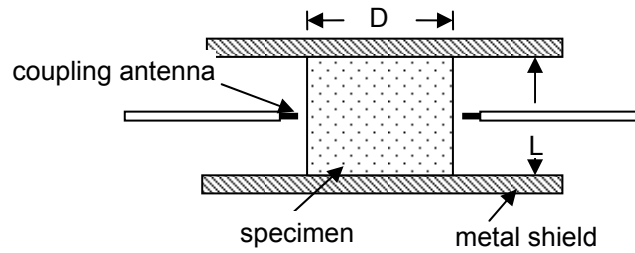

Figure 2. Dielectric constant measurement setup.

4 Results and discussions

Fig. 3 compares the theoretical curves of mixing rules to the experimental data for the ceramic-polyethylene mixtures. From the figure, we can find the curves of rules 2 and 3 match best to the physical data, followed by rules 1 and 4. From the observation in Fig. 1, the error of the rule 2 is much higher than that of the rule 3. Therefore, rule 3 is the most adequate candidate to estimate the dielectric constants of pure ceramics because it has both good curve matching and low theoretical calculation error.

To confirm the above discussions, Table 1 lists the estimated dielectric constants of pure ceramic materials from the measured dielectric constants of ceramic-polyethylene composite samples. Good estimation accuracy on dielectric constant of calcium titanate has been achieved by the rule 3 for filler concentration higher than 30%. The performance of rule 2 is much worse than that of the rule 3, although the curve matching conditions are similar. This confirm our analyses in Fig. 1, the high theoretical error of rule 2 deteriorates the measurement accuracy.

In conclusion, dielectric constant of the calcium titanate ceramic at microwave frequency can be reasonably estimated from that of the ceramic-polymer composite by choosing adequate mixture equation and filler concentration.
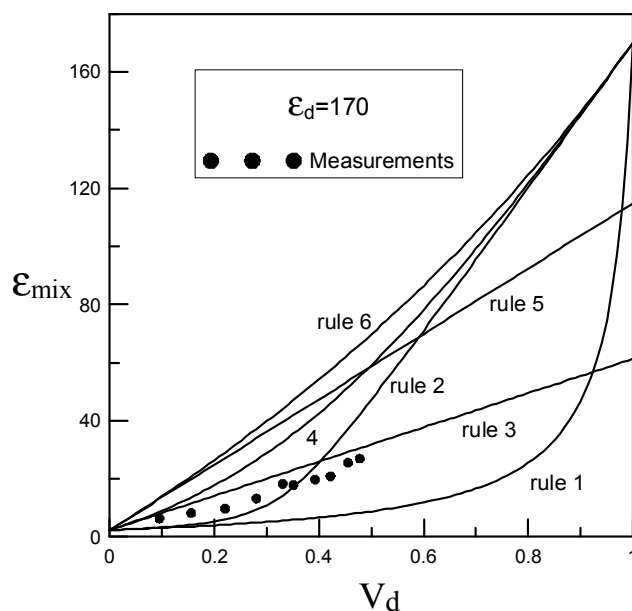
Figure 3. Comparisons of experimental data and mixture rules.

V$_d$ (%)	9.45	15.54	22.03	28.00	33.04	35.08	39.22	42.17	45.54	47.78
ε_{mix}	6.222	8.137	9.658	13.16	18.22	17.80	19.71	20.76	25.49	26.85
Rule 1	-7.03	-8.14	-9.73	-10.4	-10.8	-11.7	-13.0	-14.1	-14.6	-15.5
Rule 2	-24.5	-50.8	-161	1604	290	184	128	106	109	101
Rule 3	118	106	93.7	109.9	138	126	127	125	146	148
Rule 4	104	86.0	69.7	75.2	88.0	78.5	75.0	71.4	80.0	78.7
Rule 5	63.4	57.5	51.3	59.4	73.4	67.5	67.8	66.9	77.6	78.3
Rule 6	61.6	54.7	47.8	54.2	65.8	60.0	59.4	58.0	66.4	66.4

Table 1. Calculation of dielectric constant of pure ceramics by various mixture rules from the measured dielectric constants of composite samples. Errors less than 30% are marked with underline.

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