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Study of dielectric constants of binary composites at microwave frequency by mixture laws derived from three basic particle shapes

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ABSTRACT

Powder mixture rules derived from the filler particles with the shapes of sphere, cylinder or rod, and lamella or disk with random distributions are studied in this paper. Three ceramic powders of fillers with dielectric constants of 10, 20, and 36, respectively, are adopted. The experimental dielectric constants of ceramic dispersions in the polyethylene matrix at microwave frequency are compared to those obtained by using different mixing laws. The mixing rules are also adopted to estimate the dielectric constants of pure ceramics from the measured dielectric constants of composites with various concentrations. The theory for the error of estimation is studied. In contrast to the traditional concept of obtaining the best curve fitting of mixture rule with the experimental data, this study conclude a very important concept on the powder mixture rule, that is, the most adequate mixture law for estimating the dielectric constants of pure ceramics requires both good curve fitting and potential of less error.

1. Introduction

Starting from 1890's, permittivities of heterogeneous mixtures have been investigated by several researchers. The major application of mixture laws is often to calculate the permittivity of a ceramic material from the measurements on powder. Many theories and empirical formulas have been proposed and developed [1–18]. These studies are generally focused on obtaining the best agreement between the experimental values with the theoretical curves of various mixing laws, that is, accurate calculation of the dielectric properties of the composite samples from the mixture rules. It has been believed that this is the only or the most important goal for the study of mixture laws. However, to obtain the permittivity of a ceramic material from the powder, we need to calculate the permittivities of pure ceramic from the measurements of composite samples. The question is whether a mixture rule having the best curve matching with experimental values can guarantee the highest accuracy on the estimation of permittivities of pure ceramic or not. The answer seems negative. Based on our theoretical study, a mixture rule having larger differences with the physical values than the other rules does not mean higher calculation errors on estimating the dielectric constants of pure ceramics. Actually, various theoretical errors in the individual rules dramatically influence the accuracy of estimating the dielectric properties of a pure ceramic. We are not going to deny the importance of curve matching of mixture rules with the experimental results, but to emphasize that both the curve matching and the analyses of theoretical error are very important for mixture laws.

Powder mixture rules derived from various filler particle shapes have been proposed. The ellipsoidal shape fillers with random distribution are the one often studied. One of the most widely adopted special cases of the ellipsoidal shape is sphere, where the depolarizing factor is taken as 1/3 [1,2]. In addition to the spherical shape, the concept of ellipsoids can also be utilized to derive the expressions of long prolate spheroids (cylinders, needles, or rods). In contrast to the prolate shape, the mixture rules of fillers with flatly extended oblate spheroids (lamellae, flakes, or disks) were also published.

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Powder mixture rules of dielectric constant derived from the above three basic kinds of filler shapes (spheres, prolate spheroids, and oblate spheroids) with random distribution are investigated in this paper. The measurement frequency of this article is in the microwave range. The curve matching condition with our experimental results and analyses of theoretical error, as well as the physical error of estimating the dielectric constants of pure ceramics are studied. Polyethylene powder was chosen as the matrix material. Three kinds of ceramic powders with dielectric constants of 10, 20, and 36 were adopted as the filler materials. The dielectric constants of the three kinds of ceramic–polyethylene composites with various compositions are measured at microwave frequency. The experimental results are compared with the theoretical curves of the mixing laws to understand their agreement with the experimental values. The dielectric constants of composites are then converted to the dielectric constants of pure filler medium by these mixing laws. The accuracy of estimating the dielectric constants of pure ceramics by various mixture rules is compared. The most adequate way to select the adequate powder mixture rule will be suggested.

2. Mixture rules

2.1. Mixture rules

For composite materials containing dispersed spheres, the mixture rule can be expressed as [1–4],

$$\varepsilon_{mix} - \varepsilon_m = \frac{3V_d\varepsilon_x(\varepsilon_d - \varepsilon_m)}{2\varepsilon_x + \varepsilon_d - V_d(\varepsilon_d - \varepsilon_m)} \tag{1}$$

where ε_{mix}, ε_d, ε_m, and ε_x are dielectric constants of a composite, pure dielectric filler, polymer matrix material, and the quantity to be specified, respectively. The volume percentage of the filler medium is given by V_d. Under the special case of low filler concentration V_d, we can have $\varepsilon_x = \varepsilon_m$ and the equation becomes [1,2,5–7],

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

We now turn to the other limit of high filler concentration, $\varepsilon_x = \varepsilon_{mix}$ [1,2,5–7]. Eq. (1) becomes,

$$\frac{\varepsilon_{mix} - \varepsilon_m}{3\varepsilon_{mix}} = \frac{\varepsilon_d - \varepsilon_m}{\varepsilon_d + 2\varepsilon_{mix} - V_d(\varepsilon_d - \varepsilon_m)} V_d \tag{3}$$

A modified form of Eq. (3) has shown good curve fitting with the experimental data and accurate estimation on the dielectric constants of pure ceramics [6–8],

$$\frac{\varepsilon_{mix} - \varepsilon_m}{3\varepsilon_{mix}} = \frac{\varepsilon_d - \varepsilon_m}{\varepsilon_d + 2\varepsilon_{mix}} V_d \sim \text{rule 2, spheres with high } V_d \tag{4}$$

For filler particles with long prolate spheroids, such as cylinders, needles, or rods, random distribution of filler particles leads to the following equation [9],

$$\varepsilon_{mix} - \varepsilon_m = \frac{(\varepsilon_d - \varepsilon_m)(\varepsilon_d + 5\varepsilon_x)}{3(\varepsilon_x + \varepsilon_d)} V_d \tag{5}$$

For the cases of low and high filler concentrations with $\varepsilon_x = \varepsilon_m$ and $\varepsilon_x = \varepsilon_{mix}$, respectively, we can have [1,2],

$$\varepsilon_{mix} - \varepsilon_m = \frac{(\varepsilon_d - \varepsilon_m)(\varepsilon_d + 5\varepsilon_m)}{3(\varepsilon_d + \varepsilon_m)} V_d \sim \text{rule 3, prolate spheroids with low } V_d \tag{6}$$

and,

$$1 - V_d = \frac{(\varepsilon_d - \varepsilon_{mix})(2\varepsilon_m + \varepsilon_d + 3\varepsilon_{mix})}{(\varepsilon_d - \varepsilon_m)(\varepsilon_d + 5\varepsilon_{mix})} \sim \text{rule 4, prolate spheroids with high } V_d \tag{7}$$

The mixture rule for filler with the shapes of extended oblate spheroids, such as lamellae, flakes, and disks, can be expressed as [1,2],

$$\varepsilon_{mix} - \varepsilon_m = \frac{(\varepsilon_d - \varepsilon_m)(\varepsilon_x + 2\varepsilon_d)}{3\varepsilon_d} V_d \tag{8}$$

By applying $\varepsilon_x = \varepsilon_m$ and $\varepsilon_x = \varepsilon_{mix}$, we obtain [1,2,10],

$$\varepsilon_{mix} - \varepsilon_m = \frac{(\varepsilon_d - \varepsilon_m)(\varepsilon_m + 2\varepsilon_d)}{3\varepsilon_d} V_d \sim \text{rule 5, oblate spheroids with low } V_d \tag{9}$$

and,

$$\frac{\varepsilon_{mix}}{\varepsilon_d} = \frac{3\varepsilon_m + 2V_d(\varepsilon_d - \varepsilon_m)}{3\varepsilon_d - V_d(\varepsilon_d - \varepsilon_m)} \sim \text{rule 6, oblate spheroids with high } V_d \tag{10}$$

Powder mixture rules derived from the filler shapes of sphere, prolate spheroid, and oblate spheroid with low and high V_ds in Eqs. (2), (4), (6), (7), (9), and (10) are investigated in this article. They are adopted to compare with our measured dielectric constant values of composite samples as well as to estimate the dielectric constants of pure ceramics.

2.2. Error analyses for estimating dielectric constants of pure ceramics

To evaluate the accuracy of estimating the dielectric constants of pure ceramics from the measured dielectric constants of composites by each mixing law, the theoretical errors on dielectric constants of pure ceramics ($\Delta\varepsilon_d$) due to the dielectric constant errors of individual rules ($\Delta\varepsilon_{mix}$) are derived. From Eqs. (2), (4), (6), (7), (9), and (10), we have the error for each law,

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{\varepsilon_{mix}}{\varepsilon_d}\frac{\varepsilon_m(2+V_d) + \varepsilon_d(1-V_d)}{\varepsilon_m(1+2V_d) - \varepsilon_{mix}(1-V_d)}\frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \sim \text{rule 1} \tag{11}$$

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{\varepsilon_{mix}}{\varepsilon_d}\frac{\varepsilon_m(2-3V_d) - \varepsilon_d(1-3V_d) - 4\varepsilon_{mix}}{\varepsilon_{mix}(1-3V_d) - \varepsilon_m}\frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \sim \text{rule 2} \tag{12}$$

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{\varepsilon_{mix}}{\varepsilon_d}\frac{\varepsilon_m + \varepsilon_d}{\varepsilon_m - \varepsilon_{mix} + \frac{2}{3}(\varepsilon_d + 2\varepsilon_m)V_d}\frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \sim \text{rule 3} \tag{13}$$

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{\varepsilon_{mix}}{\varepsilon_d}\frac{6\varepsilon_{mix} + (3-5V_d)(\varepsilon_d - \varepsilon_m)}{(5\varepsilon_{mix} + 2\varepsilon_d - \varepsilon_m)V_d}\frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \sim \text{rule 4} \tag{14}$$

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{3\varepsilon_{mix}}{3(\varepsilon_m - \varepsilon_{mix}) + (4\varepsilon_d - \varepsilon_m)V_d}\frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \sim \text{rule 5} \tag{15}$$

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{\varepsilon_{mix}}{\varepsilon_d}\frac{3\varepsilon_d - (\varepsilon_d - \varepsilon_m)V_d}{3(\varepsilon_m - \varepsilon_{mix}) + (4\varepsilon_d + \varepsilon_{mix} - 2\varepsilon_m)V_d}\frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \sim \text{rule 6} \tag{16}$$

From the above equations, the theoretical error may change with the volume concentration V_d and dielectric constants of both matrix and filler. To understand the variation of the computation errors ($\Delta\varepsilon_d/\varepsilon_d$), Fig. 1 shows the variations of the $\Delta\varepsilon_d/\varepsilon_d$ with respect to the volume percentage of dispersed material assuming a 10% error of mixture rule ($\Delta\varepsilon_{mix}/\varepsilon_{mix}$) and $\varepsilon_d = 30$. The dielectric constant of 2.32 for the matrix material (polyethylene) was adopted. It can be seen that the error increases with decreasing the volume concentration of the dispersed material. Therefore, to have a more accurate estimation, higher volume percentage of V_d is a better choice. Obviously, the error of rule 1 is much higher than other rules. Therefore, it is not a good choice for calculating the dielectric constants of pure ceramics unless the rule is very accurate ($\Delta\varepsilon_{mix}/\varepsilon_{mix}$ very small). It seems that mixing rule 4 has the lowest calculation error under the same $\Delta\varepsilon_{mix}/\varepsilon_{mix}$ value. The second choices are rules 5 and 6. Then are the rules 2 and 3. By keeping V_d constant and changing ε_d, Fig. 2 shows changes of $\Delta\varepsilon_d/\varepsilon_d$ with ε_d, assuming $\Delta\varepsilon_{mix}/\varepsilon_{mix}$=10% and $V_d = 0.2$. Again, it is found that mixing rule 4 has the lowest calculation errors followed by rules 5 and 6, then rules 3 and 2. However, the physical estimation accuracy will also depend on the actual $\Delta\varepsilon_{mix}/\varepsilon_{mix}$ value of the individual mixture rules. The $\Delta\varepsilon_d/\varepsilon_d$ value of each rule will depend not only on the theoretical error but also on their matching accuracy with experimental results.

3. Experimental

Ceramic materials investigated in this article are alumina and two commercial ceramics (Prosperity Dielectrics Co., Ltd.) with dielectric constants of 10, 20, and 36 at 10 GHz, respectively. Polyethylene with measured dielectric constant of 2.32 at 10 GHz was adopted as the matrix. Material characteristics are summarized in Table 1.

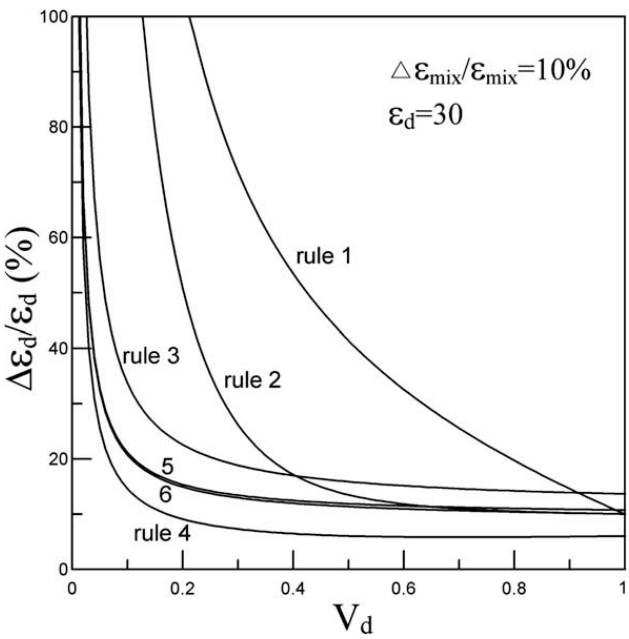

Fig. 1. Theoretical analyses on the error of estimating dielectric constant of pure ceramic vs. volume concentration of filler under the condition of $\varepsilon_d = 30$ and $\Delta\varepsilon_{mix}/\varepsilon_{mix} = 10\%$.

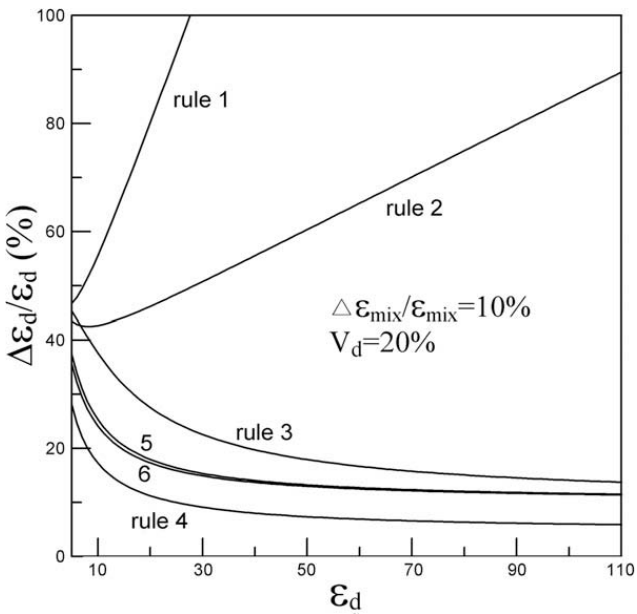

Fig. 2. Theoretical analyses on the error of estimating dielectric constant of pure ceramic vs. dielectric constant of pure filler under the condition of $V_d = 20$ and $\Delta\varepsilon_{mix}/\varepsilon_{mix} = 10\%$.

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 h. After the well mixed suspension was stirred and evaporated until almost dry, it was baked at 80 °C for 4–5 h to obtain the completely dried powder mixture. The mixed powder was then pressed using a steel die at 125 °C (the melting point of polyethylene is 115 °C) and ~7000 lb/in^2 pressure for about 0.5 h to make disk composite samples. The concentrations were calculated using the densities of polyethylene (0.915 g/cm^3) and ceramics, and the weight content of ceramics within the composite samples.

Dielectric constants of the disk-shaped composite samples are measured by the widely used Hakki and Coleman post resonance technique [18]. The main advantage for this method is that it can give very reliable measurement on dielectric constant value [19]. All the sample dimensions are designed to have the resonant frequency of desired $TE_{01\delta}$ mode at 10 GHz for measurements [19].

Table 1
Physical properties of the matrix and ceramic materials

Materials		Density (g/cm^3)	Dielectric constant (10 GHz)
Matrix	Polyethylene	0.915	2.32
Filler	Alumina ($\varepsilon_d = 10$)	3.97	10
	$\varepsilon_d = 20^*$	3.70	20
	$\varepsilon_d = 36^*$	4.50	36

* Commercial microwave ceramics. Powders are supplied by the Prosperity Dielectrics Co., Ltd., Taiwan.

4. Results and discussion

Figs. 3–5 compare the theoretical curves of the six mixture rules with the experimental data for the three ceramic–polyethylene mixtures, respectively. Table 2 lists the estimated dielectric constants of pure ceramic materials from the measured dielectric constants of composite samples. In the table, to make the comparison among various rules easier, errors less than 25% are marked with underlines. Based on the data in Table 2, three issues are worth discussing.

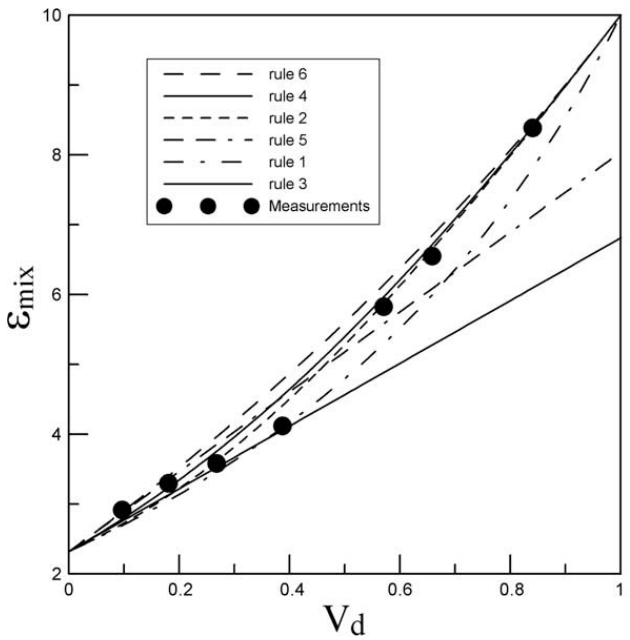

Fig. 3. Comparisons of experimental data and mixture rules for $\varepsilon_d = 10$.

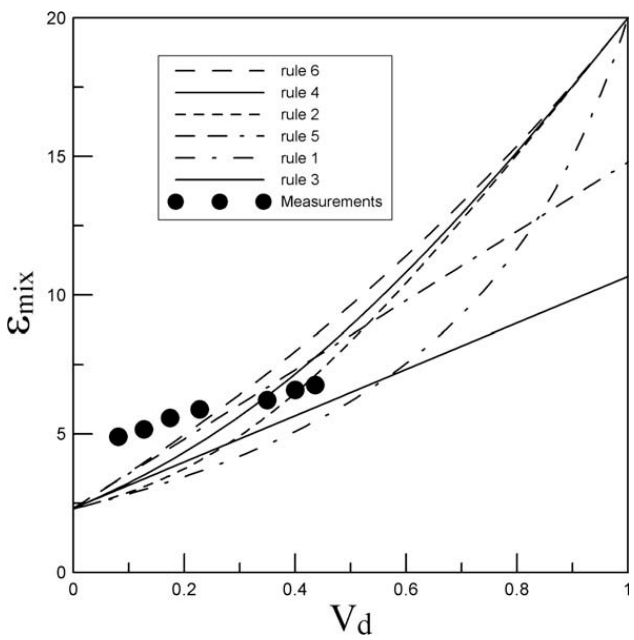

Fig. 4. Comparisons of experimental data and mixture rules for $\varepsilon_d = 20$.

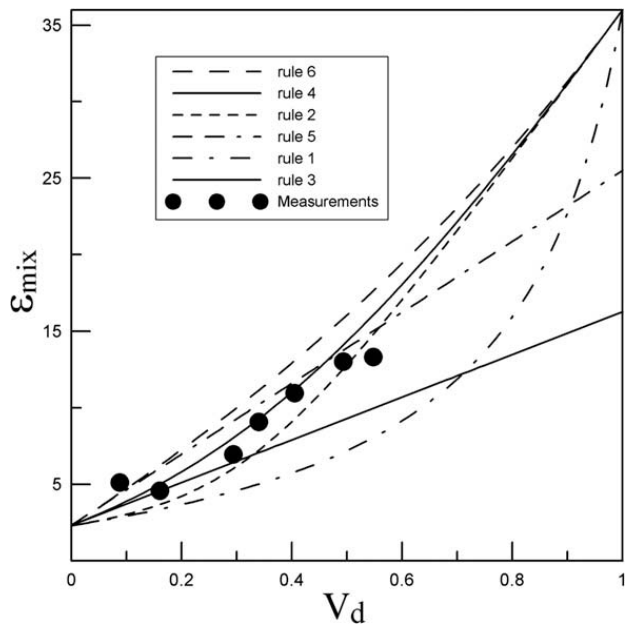

Fig. 5. Comparisons of experimental data and mixture rules for $\varepsilon_d = 36$.

First, most of the lower measurement errors fall in the region of higher filler concentration. This phenomenon agrees with our analyses of Fig. 1, which shows that the error dramatically increases upon decreasing V_d. In order to obtain better accuracy on the dielectric constants of pure ceramics from the measurements of composites, a higher filler concentration is preferred.

Second, by comparing the results of $\varepsilon_d = 10$, 20, and 36 in the table, it is found that the examples for $\varepsilon_d = 10$ have higher accuracy than that for $\varepsilon_d = 20$, and $\varepsilon_d = 36$ in estimating the dielectric constants of pure ceramics. This is conflicted with what we can see in Fig. 2, which shows the errors decrease upon increasing the dielectric constants. The reason can be explained from observation of Figs. 3–5. The experimental data match better with the theoretical curves for $\varepsilon_d = 10$ than those of $\varepsilon_d = 20$, and 36. By comparing the three figures, one can also find that the curves of various mixture rules for $\varepsilon_d = 10$ are closer with each other. Therefore, in general, the values of $\Delta\varepsilon_{mix}/\varepsilon_{mix}$ are smaller and the computation errors ($\Delta\varepsilon_d/\varepsilon_d$) are lower. Low ε_d materials work better for the estimation of dielectric constant from the composite samples due to the smaller curve differences among various rules. The contribution of good curve matching and lower $\Delta\varepsilon_{mix}/\varepsilon_{mix}$ of the low ε_d materials overcomes the theoretical higher error in Fig. 2.

Finally, we want to compare the accuracy of various mixture laws. Since the errors are dramatically increased for low filler concentrations, we will focus the analyses in the range of $V_d > 0.25$. In Table 3, the error analyses of Figs. 1 and 2 and curve fitting condition of Figs. 3–5 of the six mixture rules are listed. The final performance to estimate the dielectric constants of pure ceramics is also summarized in the table. From Table 3, we can find that rule 2 performs the best by fitting the experimental results as shown in Figs. 3–5. It does have lower errors to estimate the ε_d for examples of $\varepsilon_d = 10$ and 20. However, for $\varepsilon_d = 36$, although its curve fitting condition is the best among the six rules,

Table 2
Prediction of the dielectric constant of pure ceramics by various mixture rules from the measured dielectric constants of composite samples (data with errors less than 25% are underlined)

(a) $\varepsilon_d = 10$								
	V_d (%)	9.7	18.09	26.78	38.75	57.05	65.84	84.06
	ε_{mix}	2.91	3.29	3.58	4.12	5.82	6.55	8.38
Estimated ε_d by various mixture rules	Rule 1	38.50	18.24	12.00	10.33	12.34	11.79	10.99
	Rule 2	23.42	13.49	9.89	8.75	9.93	9.83	10.00
	Rule 3	14.54	12.49	10.70	10.48	14.20	14.92	17.00
	Rule 4	13.32	10.86	9.01	8.37	9.71	9.72	9.98
	Rule 5	10.86	9.69	8.63	8.49	10.66	11.07	12.22
	Rule 6	10.62	9.33	8.18	7.88	9.36	9.50	9.93
(b) $\varepsilon_d = 20$								
	V_d (%)	8.11	12.75	17.43	22.79	36.62	39.99	43.61
	ε_{mix}	4.90	5.17	5.58	5.89	6.44	6.59	6.76
Estimated ε_d by various mixture rules	Rule 1	−7.51	−9.89	−12.73	−18.35	−291.7	162.4	64.58
	Rule 2	−20.05	−38.35	−119.6	118.0	23.71	20.67	18.39
	Rule 3	7.18	61.29	50.36	41.33	28.39	26.74	25.31
	Rule 4	5.67	51.86	40.27	31.20	19.23	17.75	16.48
	Rule 5	6.37	34.99	29.47	24.89	18.25	17.39	16.65
	Rule 6	5.66	33.66	27.97	23.27	16.49	15.58	14.79
(c) $\varepsilon_d = 36$								
	V_d (%)	8.97	16.06	29.41	34.01	40.51	49.31	54.78
	ε_{mix}	5.11	4.57	6.95	9.07	10.95	13.00	13.32
Estimated ε_d by various mixture rules	Rule 1	−7.699	−17.35	−23.3	−19.73	−23.09	−34.22	−60.88
	Rule 2	−22.22	−378.6	53.13	58.01	47.24	37.80	31.64
	Rule 3	87.55	36.59	41.50	53.58	57.86	58.89	54.20
	Rule 4	77.67	30.05	29.07	35.08	35.15	32.78	29.02
	Rule 5	48.19	22.47	24.97	31.09	33.26	33.78	31.41
	Rule 6	46.86	21.46	22.89	28.00	29.28	28.87	26.40

Table 3
Summary of the choice of mixture rules for $V_d > 0.25$

	Choice of mixture rules	Summary
Theoretical error analyses (Figs. 1 and 2)	Errors from low to high: rules 4, (5, 6), (2, 3),1	From the calculation of pure ceramic in Table 2
Agreement with theoretical curves (Figs. 3–5)	Good to bad on curve matching: $\varepsilon_d = 10$: Rules (2, 4), (1, 5, 6), 3 $\varepsilon_d = 20$: Rules (2, 4, 5), (1, 3, 6) $\varepsilon_d = 36$: Rules 2, 4, (3, 5), (1, 6)	Errors from low to high: $\varepsilon_d = 10$: Rules (2, 4, 5, 6), 1, 3 $\varepsilon_d = 20$: Rules (2, 4, 5), 6, 3, 1 $\varepsilon_d = 36$: Rules 4, 5, (2, 6), 3, 1

Rules with similar performance are put together in the parentheses.

the errors for estimating ε_d are higher than those of rules 4 and 5, and even rule 6. The situation of rule 2 can be easily explained by the concept we have introduced before. The good performance of rule 2 for $\varepsilon_d = 10$ and 20 is because the better curve fitting overcomes the higher theoretical errors than rules 4 and 5. Obviously, for $\varepsilon_d = 36$, the curve fitting of rule 2 is still good but not good enough to compensate for the higher theoretical errors than rules 4 and 5. This confirms our former theory that good agreement with experimental data and small theoretical error are equally important for the choice of adequate mixture rule.

5. Conclusions

Dielectric constants of composite materials with various volume percentages of the three kinds of ceramic fillers and polyethylene matrix have been investigated at 10 GHz. Experimental results are compared with six mixture rules derived from three most popular particle shapes. Errors in the estimation of the dielectric constant of 100% volume percentage ceramics by the six mixture rules have been studied. Experimental results are also compared with the six mixture laws to understand the differences between the theoretical curves with the experimental values. The measured dielectric constants of composites were converted to the dielectric constants of pure ceramic materials. In order to have a more accurate estimation, a higher filler concentration is required. Both good curve matching with experimental data and small theoretical error are important for the choice of adequate mixture rule to compute the dielectric constants of ceramic materials by the measurement of powders.

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