

# Dielectric Measurements of High Dielectric Constant Materials at Microwave Frequency Using Five Well-Known Mixture Equations

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**ABSTRACT:** The dielectric constant, which is the real part of the complex permittivity, of composite materials at microwave frequencies was investigated in this study. Ceramics of titanium dioxide, calcium titanate, and strontium titanate with high dielectric constants of 100, 170, and 300, respectively, were selected. Ceramic powders were spread in the polyethylene matrix to form composite samples. The dielectric constants of the composite samples were measured to determine their matching conditions with the mathematical curves of five well-known mixture equations. These five mixture rules were then applied to estimate the dielectric constants of the three selected ceramics from the measured dielectric properties of the composite samples with various volume percentages of ceramic fillers. The mathematical equations of the potential theory errors of the five mixture rules for the dielectric constant estimation were derived and discussed. One of the five rules was selected and modified to obtain a new empirical mixture equation. This proposed empirical equation can significantly improve the accuracy of dielectric constant measurements for the selected ceramic materials. An empirical mathematical relation of the new mixing rule with the dielectric constant of the ceramic is then concluded.

## 1. INTRODUCTION

Various mixing rules have been widely adopted to calculate the dielectric constants of composite samples with varying concentrations by dispersing ceramic powders in a polymer matrix [1–10]. The development of mixture rules has been ongoing for over one hundred years. In recent years, both already existed and newly proposed equations were studied by researchers. Both low-dielectric-constant materials [11–14] and high-dielectric-constant materials [15–19] were investigated. For high-dielectric-constant materials, barium titanate was mainly studied. Regarding measurement frequencies, dielectric properties on both low frequencies (lower than 100 MHz) [15–19] and high frequencies (from GHz to THz) [11–14] were researched. Studies of high-dielectric-constant materials were mainly focused on low-frequency ranges. The investigations of high-dielectric-constant materials in high-frequency ranges, which have been less investigated in recent years, are presented in this article.

One important function of the mixture equations is to derive the dielectric constants of pure ceramics from composites with different doping percentages. For this purpose, a theoretical investigation of the potential errors of six mixture rules obtained from three fundamental particle forms of ceramic fillers has been published for ceramic examples with low dielectric constant values of no more than 40 [20]. Furthermore, the theoretical errors of the more widely adopted mixing equations were investigated [21]. Previous studies have found that not only is

there a good agreement between the theoretical curve and the physical measurement data, but the theoretical error is also critical to improve the accuracy of the dielectric constant calculation of pure ceramics. In addition to low-dielectric-constant ceramics, the application of the mixture rule to ceramic materials with higher dielectric constants, on the order of gigahertz, was investigated for titanium dioxide and calcium titanate, with real permittivities of 100 and 170, respectively [22], where the exponential and logarithmic mixing equations, in addition to the six basic equations, were studied. Although some accurate mixture equations have been suggested, it is always worthwhile to conduct further studies on the large number of published rules, especially the study of high-dielectric-constant ceramic fillers, which have been less investigated than low-dielectric-constant fillers at microwave frequencies [15–19, 23].

Therefore, five more widely adopted mixture equations were picked up to realize their reliability for dielectric properties measurement at microwave frequencies in ceramic materials having high dielectric constant characteristics. Their mathematical expressions of theoretical error are presented and studied in this article. Besides the theoretical research, experimental measurements are always required. In addition to the titanium dioxide and calcium titanate, strontium titanate has a dielectric constant of 300 at the order of gigahertz and is important for some applications at microwave frequencies [24]. A preliminary study of titanium dioxide and strontium titanate with dielectric constants of 100 and 300, according to the five rules, has been given in our conference papers [25] and [26]. However, the calcium titanate with a dielectric constant of 170

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has not been studied by the five rules. In addition to a new study about the calcium titanate, the previous results in [25] and [26] are all integrated in this article. This research, therefore, provides an overall understanding of the five popular mixture rules for high dielectric constant ceramics. This integrated study also develops a new relationship formula among the mixture rules observed from the three ceramic materials.

The powders of the above three ceramics were then used as ceramic fillers to mix with the matrix powder of polyethylene. Measurements at 10 GHz were conducted on the composite samples with different concentrations for their dielectric constant values. The mathematical curves of the five mixing rules were juxtaposed with the measurements to determine the curve matching error of each mixture equation. Furthermore, based on the measurement results and theoretical calculation errors, in addition to observing the matching error, one mixture equation was selected and modified to build a new powder mixing rule. The newly constructed mixing equation has been proven to have a very low curve matching error with the measurement results and a very low potential theory error. A modification factor was introduced in this rule, whose value changes with various ceramics. Finally, the real permittivity values of the pure ceramic materials were computed using the five selected mixture equations, as well as the newly suggested rule from the experimental dielectric constants of the composite samples. The reliability of the six mixing rules for calculating the dielectric constants of the ceramic fillers is discussed. The newly suggested mixture formula has proven to have very good calculation accuracy for the dielectric constant measurements of the three selected high-dielectric-constant ceramics.

## 2. THEORETICAL ANALYSES

### 2.1. Mixture Equations

Here, the five widely used mixing rules are introduced. The first two mixture equations are from Bruggeman's equations [1–4],

$$1 - V_d = \left( \frac{\varepsilon_d - \varepsilon_{mix}}{\varepsilon_d - \varepsilon_m} \right) \left( \frac{2\varepsilon_d + \varepsilon_m}{2\varepsilon_d + \varepsilon_{mix}} \right) \quad (1)$$

$$1 - V_d = \frac{\varepsilon_{mix} - \varepsilon_d}{\varepsilon_m - \varepsilon_d} \left( \frac{\varepsilon_m}{\varepsilon_{mix}} \right)^{1/3} \quad (2)$$

where  $\varepsilon_{mix}$ ,  $\varepsilon_d$ , and  $\varepsilon_m$  are the dielectric constants of the composite, pure dielectric filler, and polymer matrix material, respectively, and  $V_d$  is the volume percentage of the filler medium. Equations (1) and (2) are referred to as Bruggeman's symmetrical and asymmetrical mixture rules, respectively.

The third popular mixing formula is the Maxwell-Garnett rule. This equation is one of the originally suggested mixing rules, which was first proposed by Rayleigh in 1892 [1, 2, 5–8],

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

The fourth was suggested by Aspnes, who extended the Lorenz-Lorentz equation [5, 9],

$$\frac{\varepsilon_{mix} - 1}{\varepsilon_{mix} + 2} = V_d \frac{\varepsilon_d - 1}{\varepsilon_d + 2} + (1 - V_d) \frac{\varepsilon_m - 1}{\varepsilon_m + 2} \quad (4)$$

Finally, an empirical mixing formula was chosen, which was proposed by Wagner [3, 10],

$$\frac{\varepsilon_{mix} - \varepsilon_m}{3\varepsilon_m} = V_d \frac{\varepsilon_d - \varepsilon_m}{\varepsilon_d - 2\varepsilon_m} \quad (5)$$

The differences in the measurement results with the function curves of mixing Equations (1)–(5) were investigated. The theoretical errors of these equations for a high-dielectric-constant filler were then studied.

### 2.2. Analyses of Theoretical Errors

The theoretical calculation errors in the dielectric constants of pure ceramics ( $\Delta\varepsilon_d$ ) because of the errors in the composite samples of individual rules ( $\Delta\varepsilon_{mix}$ ) are [21],

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{\varepsilon_{mix}}{\varepsilon_d} \frac{\varepsilon_d(3 - V_d) + \varepsilon_m V_d}{3\varepsilon_m - \varepsilon_{mix}(3 - V_d) + V_d(4\varepsilon_d - 2\varepsilon_m)} \frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \quad (6)$$

for the first Bruggeman's mixture rule of Equation (1);

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{1}{3} \frac{\varepsilon_{mix}}{\varepsilon_d} \frac{3\varepsilon_m^{1/3} - \varepsilon_{mix}^{-2/3}(\varepsilon_m - \varepsilon_d)(1 - V_d)}{\varepsilon_m^{1/3} - \varepsilon_{mix}^{1/3}(1 - V_d)} \frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \quad (7)$$

for the second Bruggeman's mixture rule of Equation (2);

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{\varepsilon_{mix}}{\varepsilon_d} \frac{V_d(\varepsilon_d - \varepsilon_m) - (\varepsilon_d + 2\varepsilon_m)}{(\varepsilon_{mix} - \varepsilon_m) - V_d(\varepsilon_{mix} + 2\varepsilon_m)} \frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \quad (8)$$

for the Rayleigh's mixture rule of Equation (3); and

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{\varepsilon_{mix}}{\varepsilon_d} \frac{\varepsilon_d(1 - V_d) + 2 + \varepsilon_m V_d}{\varepsilon_m + 2V_d - \varepsilon_{mix}(1 - V_d)} \frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \quad (9)$$

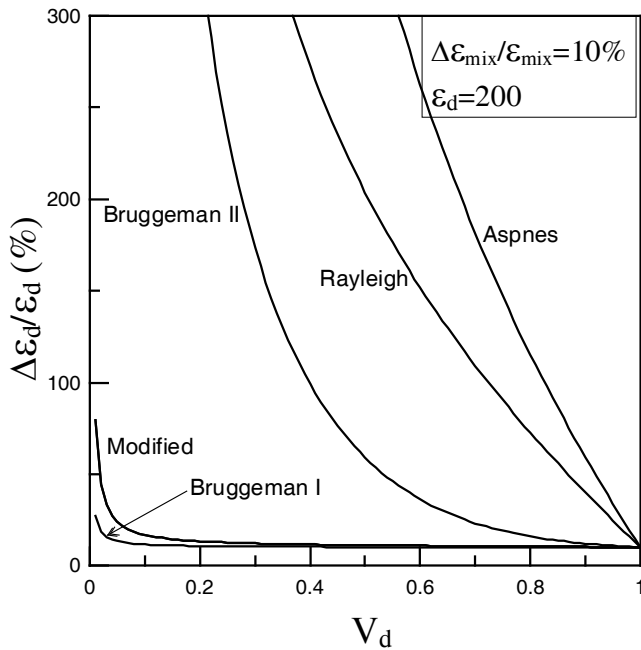
$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{\varepsilon_{mix}}{\varepsilon_d} \frac{\varepsilon_d + 2\varepsilon_m}{\varepsilon_m(3V_d + 1) - \varepsilon_{mix}} \frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \quad (10)$$

for the Aspnes' and Wagner's mixture rules, respectively.

In Equations (6) to (10), the potential theory errors vary with the concentration  $V_d$  and dielectric constants of filler and matrix,  $\varepsilon_d$  and  $\varepsilon_m$ . The relation of the calculation errors ( $\Delta\varepsilon_d/\varepsilon_d$ ) with  $V_d$ , in Equations (6) to (9), is presented in Figure 1, with the assumption of  $\Delta\varepsilon_{mix}/\varepsilon_{mix} = 10\%$ ,  $\varepsilon_d = 200$  as an example. The  $\varepsilon_m$  value is 2.32 for polyethylene matrix material. Similar relations exist for  $\Delta\varepsilon_{mix}/\varepsilon_{mix}$  other than 10% and  $\varepsilon_d$  other than 200. The error curve of Equation (10) is not in the figure because all the errors are over 100%. Based on the results of Figure 1, Equation (1) will be selected for modification due to its low theoretical error, and the modification rule will be described later. The error curve of this modified rule is also presented in Figure 1. Theoretically, the calculation error reduces with increasing the volume composition  $V_d$

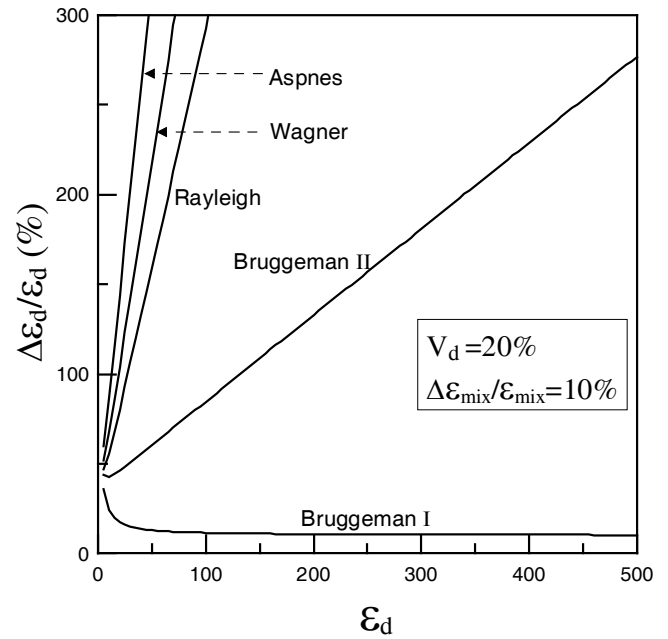
**TABLE 1.** Characteristics of the polyethylene matrix and ceramic fillers.

Materials		Density (g/cm <sup>3</sup> )	Dielectric constant (5–10 GHz)
Matrix	Polyethylene	0.915	2.32
Filler	TiO <sub>2</sub>	4.232	100
	CaTiO <sub>3</sub>	3.982	170
	SrTiO <sub>3</sub>	5.180	300



**FIGURE 1.** Theoretical error of calculating the dielectric constant of pure ceramic vs. volume concentration of filler under the condition of  $\epsilon_d = 200$  and  $\Delta\epsilon_{mix}/\epsilon_{mix} = 10\%$ . For the modified rule, the modification factor  $x = 0.175$  was obtained from Equation (13).

of the filler material, as can be seen in the figure too. It is clear that only the first Bruggeman’s rule exists as an acceptable calculation error. The errors of the other four mixing rules are too high to have a reasonable prediction of the dielectric constants of pure ceramic materials unless their theoretical curves match very well with the physical values ( $\Delta\epsilon_{mix}/\epsilon_{mix}$  very low). The variations of  $\Delta\epsilon_d/\epsilon_d$  and the dielectric constant of filler material,  $\epsilon_d$ , are presented in Figure 2, with the assumption of  $\Delta\epsilon_{mix}/\epsilon_{mix} = 10\%$  and  $V_d = 0.2$ . Similar variations as Figure 2 exist for  $\Delta\epsilon_{mix}/\epsilon_{mix}$  other values than 10% and  $V_d \neq 0.2$ . Again, except for the first Bruggeman’s rule, the theoretical error increases dramatically with increasing  $\epsilon_d$  for the other rules. Therefore, the other rules are not adequate for the high dielectric constant material, such as strontium titanate. However, as we mentioned before, the physical estimation accuracy will also rely on the real  $\Delta\epsilon_{mix}/\epsilon_{mix}$  condition of each mixing equation, which will be obtained by experimental results.

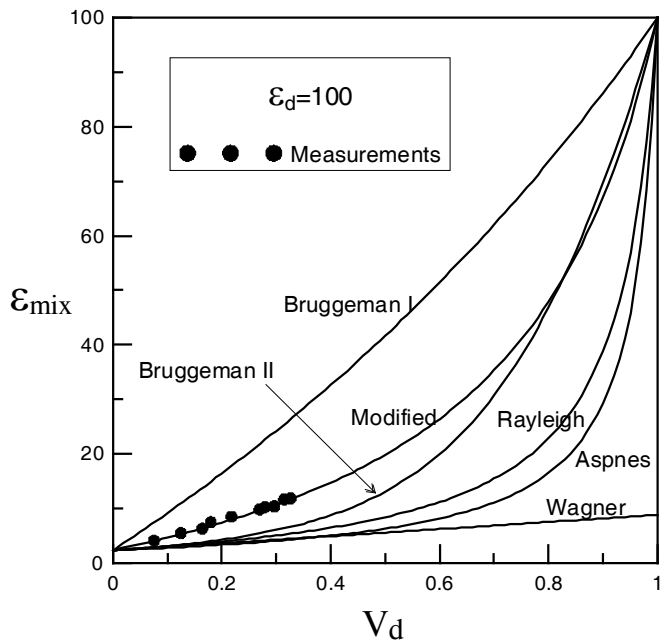


**FIGURE 2.** Theoretical error of calculating dielectric constant of pure ceramic vs. dielectric constant of pure filler under the condition of  $V_d = 20\%$  and  $\Delta\epsilon_{mix}/\epsilon_{mix} = 10\%$ .

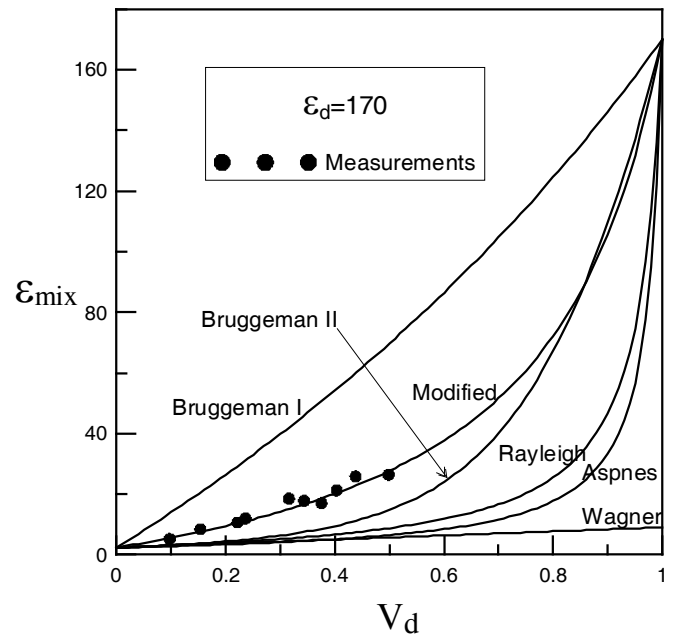
### 3. EXPERIMENTS AND MODIFICATION OF MIXTURE EQUATION

#### 3.1. Experiments

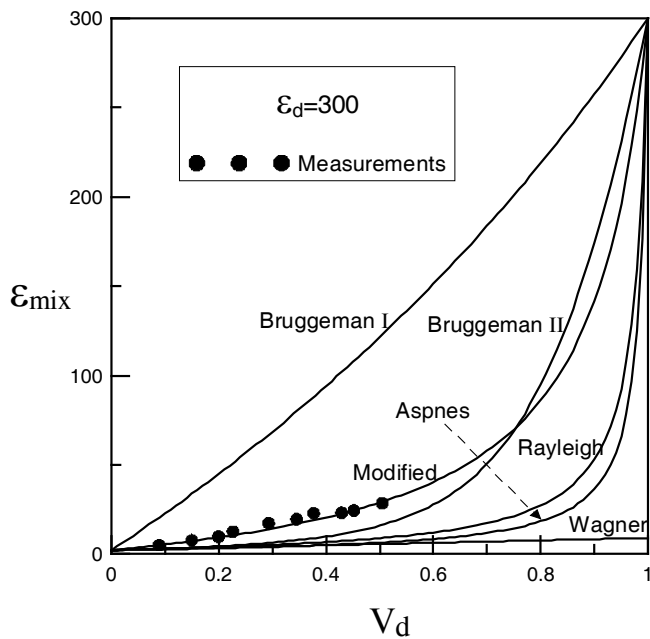
Titanium dioxide, calcium titanate, and strontium titanate ceramics have dielectric constants of 100, 170, and 300, respectively, at microwave frequencies spanning from 5 to 10 GHz, where the dielectric constant remains stable in this microwave range [20]. The matrix material polyethylene has a low dielectric constant of 2.32 at the same frequency. The physical properties of the matrix and filler materials are listed in Table 1. The fabrication methods for the composite samples have been introduced elsewhere [20]. The dielectric constants of the composite samples were measured using the Hakki-Coleman post-resonance technique at 10 GHz [27]. The cylinder-shaped sample was placed between and contacted with two parallel metal plates. The transmission coefficient of the resonant signal of the TE<sub>011</sub> mode was measured using the HP N5230A Network Analyzer or the calculation of the dielectric constant. This resonant method can provide an accurate dielectric constant, but it



**FIGURE 3.** Comparisons of experimental data and mixture rules of the titanium dioxide/polyethylene composites [18]. For the modified curve,  $x = 0.25$  in Equation (11).



**FIGURE 4.** Comparisons of experimental data and mixture rules of the calcium titanate/polyethylene composites. For the modified curve,  $x = 0.2$  in Equation (11).



**FIGURE 5.** Comparisons of experimental data and mixture rules of the strontium titanate/polyethylene composites [19]. For the modified curve,  $x = 0.1$  in Equation (11).

can only measure at the resonant frequency of the sample. The dielectric property measurement technique has been discussed elsewhere [27, 28].

Figures 3, 4, and 5 compare the mathematical curves of the five mixing rules and the modified rule to the experimental data of composites made from titanium dioxide, calcium titanate, and strontium titanate, respectively. Figures 3 and 5 refer to our conference papers [25, 26]. Figure 4 shows the results of our

new experiments. The curves of the modified rule matched the experimental results better than those of the other five rules in equations (1) to (5). The derivation of the modified rule is presented in the next section. From Figures 3 to 5, the  $\Delta\epsilon_{mix}/\epsilon_{mix}$  values for the curves of Equations (1) to (5) are far from the acceptable  $\Delta\epsilon_d/\epsilon_d$  values. A modified mixing equation based on the existing five mixture rules was then developed and suggested.

### 3.2. Derivation of Modified Mixture Equation

As shown in Figures 3–5, no good matching is found between the function curves and the measurement data for any mixture equation. A modification to the existing rules is proposed to improve the matching condition with the physical results. We attempted to choose an adequate mixing rule from the five rules for modification. From Figures 1 and 2, the first Bruggeman’s rule of Equation (1) is chosen as the rule to be modified because its mathematical curves have much lower errors on  $\Delta\epsilon_d$  than the other four rules for the same  $\Delta\epsilon_{mix}$  value, although its matching conditions are not the best in Figures 3 to 5. A modification factor is then inserted into Equation (1) as follows [25, 26]:

$$1 - V_d = \left( \frac{\epsilon_d - \epsilon_{mix}}{\epsilon_d - \epsilon_m} \right) \left( \frac{x\epsilon_d + \epsilon_m}{x\epsilon_d + \epsilon_{mix}} \right) \quad (11)$$

A coefficient designated as  $x$  is inserted in front of the two  $\epsilon_d$ ’s in the second bracket. The modification coefficient  $x$  is an empirical value that is equal to 2 in the unmodified first Bruggeman rule. This modification must obey two basic conditions:  $\epsilon_{mix} = \epsilon_d$  for  $V_d = 1$  and  $\epsilon_{mix} = \epsilon_m$  for  $V_d = 0$ . Based on the observations in Figures 3–5, to obtain the best curve fitting condition,  $x = 0.25, 0.2,$  and  $0.1$  were chosen in Figure 3

**TABLE 2.** Estimation of dielectric constant of three ceramic materials by various mixture rules from the measured dielectric constants of ceramic-polyethylene composite samples.

(a) Estimation of  $\epsilon_d$  by various mixture rules for titanium dioxide ( $\epsilon_d = 100$ ) [18].

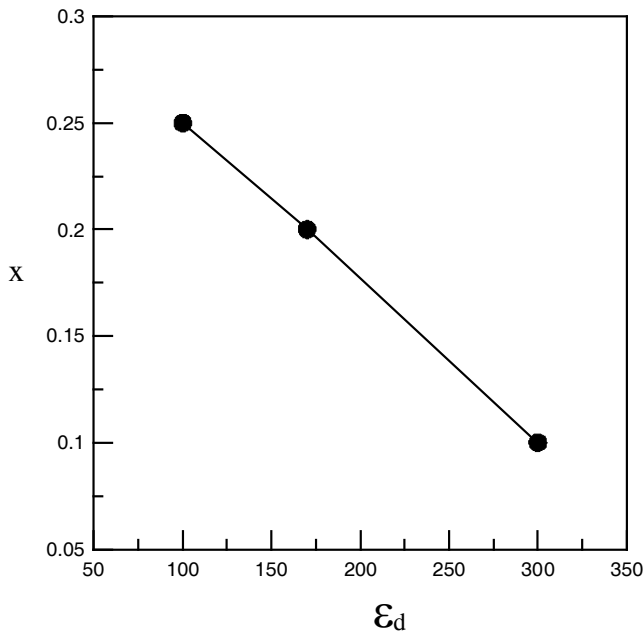
$V_d$ (%)	0.0757	0.1248	0.1642	0.1800	0.2179	0.2694	0.2796	0.2964	0.3145	0.3267
$\epsilon_{mix}$	4.114	5.463	6.249	7.481	8.485	9.833	10.233	10.369	11.642	11.840
Modified ( $x = 0.25$ )	104.72	106.86	98.74	116.35	110.56	103.31	103.81	97.61	104.99	101.74

(b) Estimation of  $\epsilon_d$  by various mixture rules for calcium titanate ( $\epsilon_d = 170$ ).

$V_d$ (%)	0.0970	0.1531	0.2209	0.2359	0.3157	0.3453	0.3751	0.4031	0.4379	0.4984
$\epsilon_{mix}$	5.155	8.369	10.602	11.974	18.464	17.780	16.690	21.206	25.780	26.378
Modified ( $x = 0.2$ )	152.27	198.02	174.96	188.74	217.80	184.58	161.99	178.78	196.38	161.94
Bruggeman I	43.71	57.57	53.49	57.96	70.10	61.28	55.62	62.39	70.22	62.03
Bruggeman II	-13.41	-17.76	-25.69	-27.53	-41.37	-49.63	-65.48	-72.83	-88.72	-181.27
Rayleigh	-8.02	-8.00	-9.29	-9.28	-10.26	-11.41	-13.09	-13.04	-13.62	-16.93
Aspnes	-3.38	-3.52	-4.17	-4.22	-4.86	-5.36	-6.05	-6.21	-6.62	-8.12
Wagner	-6.71	-6.05	-6.15	-6.00	-5.68	-5.85	-6.08	-5.79	-5.62	-5.75

(c) Estimation of  $\epsilon_d$  by various mixture rules for strontium titanate ( $\epsilon_d = 300$ ) [19].

$V_d$ (%)	0.0886	0.1496	0.1995	0.2262	0.2930	0.3450	0.3767	0.4287	0.4517	0.5046
$\epsilon_{mix}$	5.143	7.788	9.826	12.672	17.277	19.565	22.820	23.170	24.452	28.531
Modified ( $x = 0.1$ )	290.4	327.0	318.6	379.8	392.2	357.8	374.2	307.4	298.7	290.7



**FIGURE 6.** Relationship of  $\epsilon_d$  and the modification factor  $x$ .

(titanium dioxide composites), Figure 4 (calcium titanate composites), and Figure 5 (strontium titanate composites), respectively. The modified equations and experimental results are in good agreement in all three figures. The theoretical error is then derived using the differential method as follows:

$$\frac{\Delta\epsilon_d}{\epsilon_d} = \frac{\epsilon_{mix}}{\epsilon_d} \frac{(1-x-V_d)\epsilon_d + V_d\epsilon_m}{2xV_d\epsilon_d + (V_d-1-x)\epsilon_{mix} + (1-x-xV_d)\epsilon_m} \frac{\Delta\epsilon_{mix}}{\epsilon_{mix}} \quad (12)$$

The curve is also shown in Figure 1 for  $x = 0.175$  and  $\epsilon_d = 200$ . As shown in the figure, except for the first Bruggeman's equation, the theoretical error of this new rule is much better than the other four mixture equations, and its  $\Delta\epsilon_d/\epsilon_d$  value is under a reasonably low enough condition. The relationship between  $\epsilon_d$  and the modification factor  $x$  is shown in Figure 6. A linear relationship is observed in the figure between the  $\epsilon_d$  and  $x$ ,

$$200x = 65 - 0.15\epsilon_d \quad (13)$$

Therefore, the modification factor of other ceramic materials can be easily obtained from Figure 6.

#### 4. RESULTS AND DISCUSSIONS

To verify the reliability of the proposed new mixture equation, the dielectric constants of pure titanium dioxide, calcium titanate, and strontium titanate derived from the experimental results of the composite specimens using the new equation are presented in Table 2, along with those of the five popular mixing rules. This newly proposed mixture rule has shown accurate measurements of the dielectric constants of titanium dioxide, calcium titanate, and strontium titanate (expected  $\epsilon_d = 100$ , 170, and 300, respectively). The very high errors of the second Bruggeman, Rayleigh, Aspnes, and Wagner equations are also observed in Table 2(b), which agrees with our previous discussion. High errors were also found for these four rules in references [25] and [26]. The error of the first Bruggeman's rule is lower than that of the other four mixing equations, as in [25] and [26], but its accuracy is still much lower than that of the new mixture equation. One important observation can be made between the two Bruggeman mixing equations. In Figures 3 to 5, the second Bruggeman's equation has a much better curve matching to the experimental data than that of the

first Bruggeman's equation, but finally, the first equation has a higher measurement accuracy in Table 2. This is because the higher potential theory accuracy of the first Bruggeman's rule, as shown in Figures 1 and 2, overcomes its deficit of higher mismatching error. This phenomenon demonstrates the importance of the theoretical error analyses in these two figures for the powder mixture equations.

The  $V_d$ s of most of the samples are less than 50%. There are two reasons for this. First, the sample will be more difficult to shape as the concentration of polyethylene is decreased. Second, porosity may exist inside the samples, which will influence the accuracy of the dielectric constant measurement. The effect of porosity deteriorates if the concentration of ceramic powder is increased. The related discussions have also been presented in [15, 16].

In conclusion, five widely adopted mixture rules were selected for the microwave measurements of the dielectric properties of titanium dioxide, calcium titanate, and strontium titanate ceramics using ceramic-polyethylene composite samples with different volume concentrations at 10 GHz. The potential theory errors in the calculation of the dielectric constant of ceramic fillers using different mixing equations for high ceramic dielectric constants were analyzed. Based on the theoretical analysis and experimental results, a modified new mixture rule was proposed. The experimental results from various mixture rules were compared to verify the reliability of the modified mixture equation. Accurate experimental measurements of the dielectric constants of three pure ceramic materials were realized using the proposed new mixture rule.

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