Extended Mixing Rule to Complex Permittivity

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ABSTRACT

Various types of equation for mixing rule on permittivity of mixture have been proposed, but none of these is not perfect because of the inconsistency between the actual geometrical configuration and the basic model for calculation. Serial model and parallel model are lower and upper extremes of mixing manner, the apparent permittivity of any other type of mixture stay between these two extreme states. For the random mixture of the stumpy fine particles, customarily the logarithmic mixing rule has been applied. But, the logarithmic mixing rule does not give the proper value of permittivity in low or high mixing rate of constituent. The author proposed the new mixing rule that gives better consistency with measured value in whole mixing range compared to the logarithmic rule. In this paper, a desirable refinement on the equation proposed in the previous paper is made to adapt to the configuration image of actual compound and then the equation has been expanded to the complex permittivity to apply the mixing rule on the dissipative materials cases.

Key words : Complex perimittivity, TiO$_2$ ceramics, Mixing rule

1. Introduction

As the most flexibly applicable mixing rule for a mixture, the next formula is well known

$$\varepsilon^\alpha = \sum_{i=1}^{N} V_i \varepsilon_i^\alpha \tag{1}$$

where ε, ε_i, V_i and N are the apparent permittivity of mixture, permittivity and volume fraction of the i-th constituent and number of constituents, respectively. Depending on the value of parameter α (real number, $\alpha = -1$ to $+1$); equation (1) corresponds to the typical models of mixing rule as follows;
when

$\alpha = +1$	parallel model
0	logarithmic mixing rule
-1	serial model

For the homogeneous and random mixture of two constituents, logarithmic mixing rule has been most commonly applied without particular consideration in many cases. Logarithmic mixing rule would give fairly good estimated value of apparent permittivity in the volume fraction range between 0.3 and 0.5, but it does not predict the apparent permittivity in good agreement with measured value in lower and higher mixing rate ranges. It was confirmed from

the consideration of several experimental data and Finite Element Method Simulation(FEM) that; in the lower mixing range, the specific dielectric constant of mixture shows smaller value compared with that by logarithmic rule, i.e. the value of dielectric constant of mixture deviates toward to serial model side; on the other hand in the higher mixing range, it shows larger value of dielectric constant than that by logarithmic mixing rule, i.e. the mixing rule deviates toward parallel model side. Based on this results, the author have proposed the new mixing rule; in the eq. (1), exponent α takes negative value between $V_x = 0$ and V_c, (V_c is a certain volume fraction where the mixing rule moves over from serial like model to parallel like model), and over this critical fraction, α takes positive value (i.e. parallel like). The following equation gives better agreement with experimental data in the whole mixing range by replacing the parameter α in equation (1) with new one, $(V_x - V_c)$;[1]

$$\varepsilon_r^{(V_x - V_c)} = V_x \varepsilon_{r1}^{(V_x - V_c)} + (1 - V_x) \varepsilon_{r2}^{(V_x - V_c)} \tag{2}$$

or explicit form with respect to apparent permittivity

$$\varepsilon_r = exp \left[ln \left\{ \frac{V_x \varepsilon_{r1}^{(V_x - V_c)} + (1 - V_x) \varepsilon_{r2}^{(V_x - V_c)}}{(V_x - V_c)} \right\} \right] \tag{3}$$

where, ε_r, ε_{r1}, ε_{r2}, V_x, V_c are specific permittivity of mixture, material 1, material 2, volume fraction of material 2 and critical volume fraction rate.

But, in an actual mixture, at the both extreme ends of mixing rate, the mixture is simply composed with material 1 or material 2 itself, so that $(V_x - V_c)$ should be equal to -1 or $+1$ for $V_x = 0$ or 1. These requirements were not satisfied

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with the equation in the previous paper.

In this work, to eliminate this discrepancy between model and equation, index $(V_x - V_c)$, which correspond to the deviation of volume mixing rate from critical one, was normalized by V_c between $V_x = 0$ and V_c, and by $(1 - V_c)$ between $V_x = V_c$ and 1. With this modification, the inconsistency between model and equation was removed, and the values of αs are -1, 0 and $+1$ at $V_x = 0$, V_c and 1, respectively.

$$\alpha = (V_x - V_c)/V_c \qquad \text{for} \quad V_x = 0 \text{ to } V_c$$
$$\alpha = (V_x - V_c)/(1 - V_c) \quad \text{for} \quad V_x = V_c \text{ to } 1$$

Next, the equation has been formally expanded into the complex permittivity to apply the mixing rule on the mixture of dissipative or conductive materials.

$$\varepsilon^*_r = exp[ln\{V_x\varepsilon^{*\alpha}_{r1}+(1-V_x)\varepsilon^{*\alpha}_{r2}\}/\alpha]$$
$$\varepsilon^*_{r1} = \varepsilon'_{ri}-j\sigma_i/\varepsilon_0 = \varepsilon'_{ri}-j\varepsilon_{ri}'' \qquad (4)$$

where ε^*_{ri}, ε'_{ri}, ε''_{ri}, ε_0, and σ_i are the complex specific dielectric constant, real- imaginary- part of specific dielectric constant, dielectric constant of free space and electric conductivity, respectively.

2. Data and Consideration

Fig. 1 shows the specific dielectric constant data for the TiO_2 ceramics containing several amounts of pores together with the simulated curves by serial, parallel, logarithmic, and new equations. As it is seen in this figure, the measured specific dielectric constant values deviate toward parallel model and new equation does fit best with experimental data. Similar good agreements were observed with other data reported by several researchers.[2-11] Fig. 2 shows similar data for the pressed $CaTiO_3$ fine powder $(\varepsilon_r \cong 150)$, that is random mixture of $CaTiO_3$ and air. As it is seen, at around 0.35 of mixing fraction, the measured permittivity values are close to those calculated by logarithmic rule, but these deviate from logarithmic rule in the lower and higher mixing ranges. Fig. 3(a), (b), and (c) shows the relation between the volume fraction and the real, imaginary parts and absolute values of the apparent specific permittivity for the virtual mixture of Rutile powder and air calculated by new equation, logarithmic, parallel, serial models and simulated by using FEM as well. Fig. 4(a) and (b) shows the image of electric field distributions, and (c) shows the current distri-
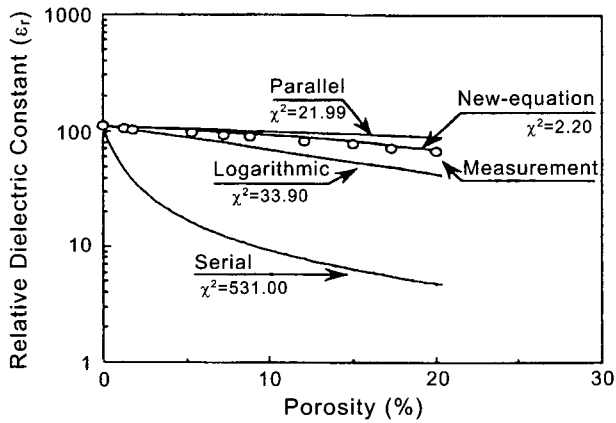

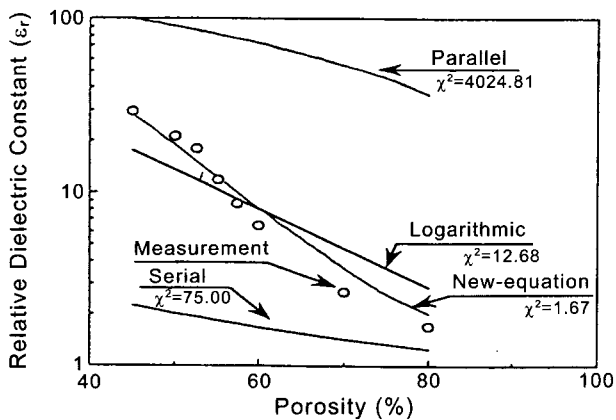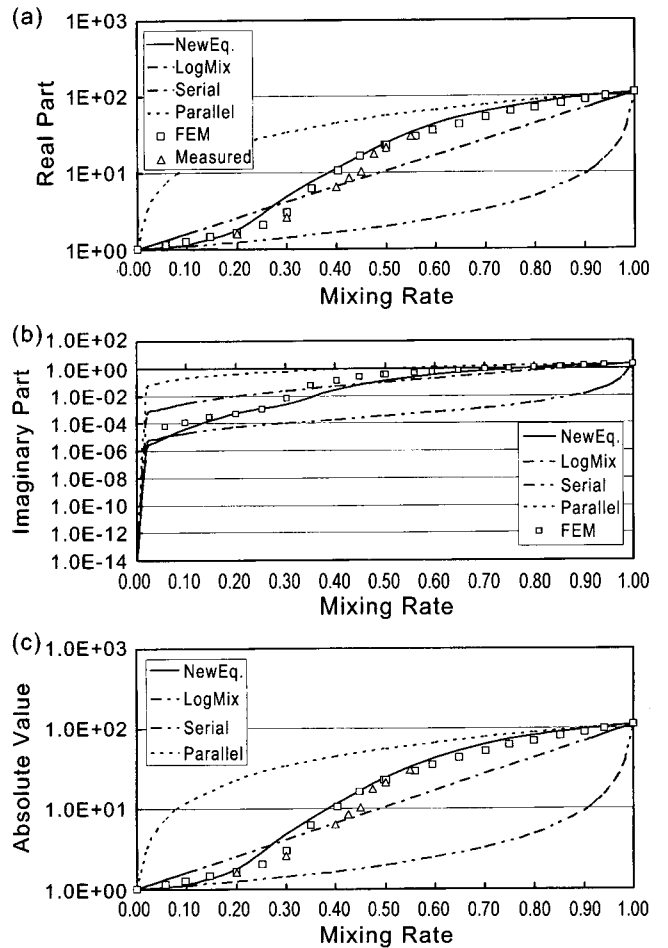Fig. 1. Apparent relative dielectric constant vs. porosity content for TiO_2 ceramics.

Fig. 2. Apparent relative dielectric constant vs. powder content for the pressed plate of $CaTiO_3$ powder and air.

Fig. 3. Complex relative dielectric constant of virtually designed mixture of TiO_2 powder and air; (a) real part, (b) imaginary part, and (c) absolute value.
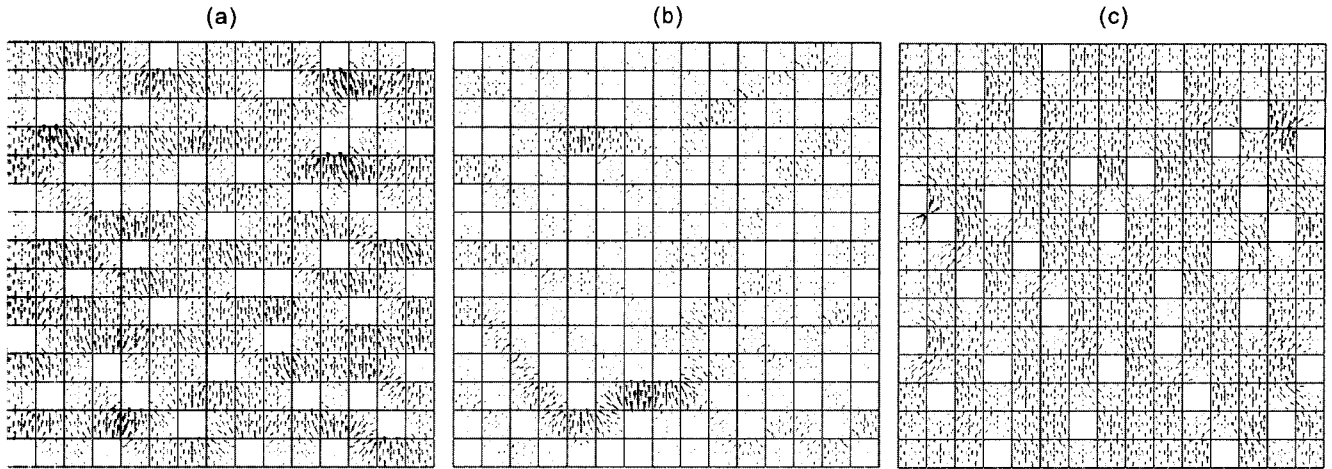
Fig. 4. Distributions of field pattern for 0.2, 0.4 and 0.8 volume fraction mixture of TiO$_2$ powder and air; (a), (b) electric field, and (c) imaginary (displacement current) distributions.

bution image pattern in the 20, 40 and 80-vol% mixtures of TiO$_2$ and air, respectively, which typically correspond to low, medium and high concentrations. As it is seen in these figures; for the low concentration mixture, electric field distribution pattern looks like to that of series connection of cells and mixing rule is close to the serial model; on the contrary, for the high concentration mixture the imaginary flux (or current) distribution pattern seems to be similar to that of parallel connection of cells and mixing rule close to parallel model; and then in the mid concentration range, these are complex of both cases. Fig. 5(a), (b), and (c) shows the components of complex permittivity for the mixture of highly dissipative (or rather semi conductive) powder and insulating elastomer with medium dielectric loss (approximately 0.1% of tan δ). Point plots of complex permittivity for the absorbing rubber compounds mixed with carbon black powder measured at medium frequency (1 MHz) and at microwave frequency (9.4 GHz) are also shown. The permittivity measured at medium frequency (probably from low frequency up to few hundreds MHz) sharply increases with increase of carbon contents at around 10 vol%. This appearance will be due to the high connectivity structure between carbon black powders, which lead to high electrical conduction. The scheme of this result seems to be close to that of the percolation phenomena. But at very high frequency (over 1 GHz), electrical conductivity due to the point contacts between carbon black powders will not play important role compared to the case at dc or lower frequency, and then apparent dielectric property seems to approach to the newly proposed mixing rule or logarithmic mixing rule rather than percolation phenomena. In fact, the electrical and/or magnetic properties of materials show very specific dependence on frequency by material and structure. Without knowing the very high frequency characteristics of each constituent and structure, it is difficult to estimate the permittivity of mixture. In the calculation for Fig. 5, we assumed that; for carbon black powder, $\varepsilon_r' = 1.0$, $\sigma = 2 \times 10^4$ S/m; and for rub-
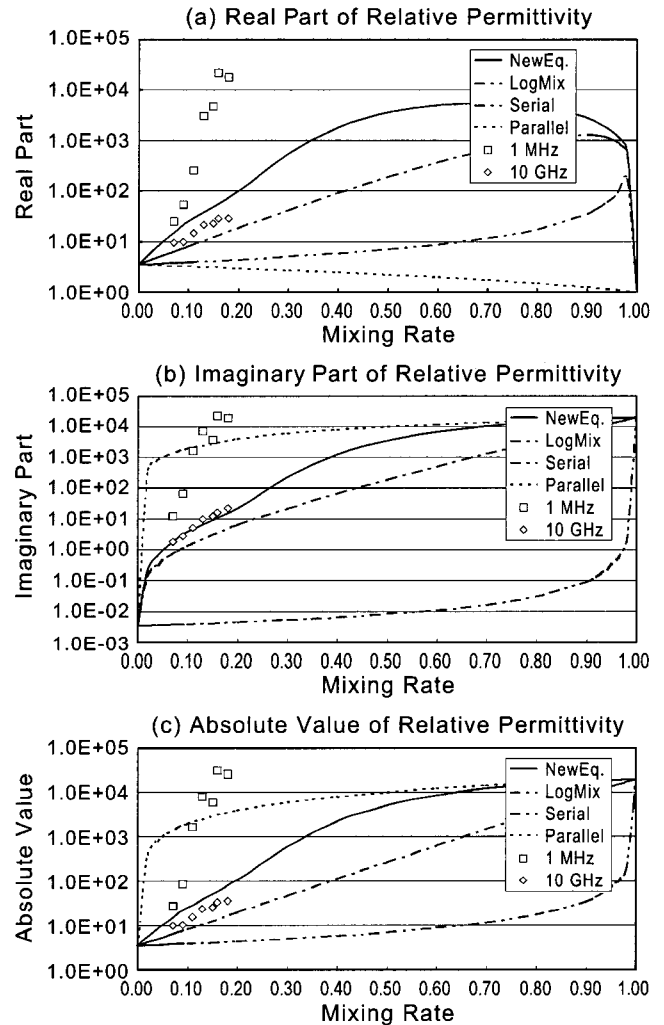

Fig. 5. Complex relative dielectric constant for virtually designed mixture of carbon black and elastomer, and measured data for conductive powder and butyl-rubber compound for EMC absorber are point plotted; (a) real part, (b) imaginary part, and (c) absolute value.

ber $\varepsilon_r' = 3.5$, $\sigma = 1 \times 10^{-14}$ S/m and no specific structure.

For the mixture of a conductive or semi-conductive powder and insulating elastomer, the apparent dielectric characteristics calculated by the new equation show the similar tendency to the relation between conductivity and conductive powder concentration of a percolation phenomena. In the percolation phenomena under the dc electric field, the electric current path is created by the real contact between conductive particles. But, in case of ac phenomena, especially at higher frequency, a dielectric displacement of elastomer creates the virtual current path between conductive particles through the suspending insulator without actual contact, and then the mixture of insulator and conductive powder will show the similarity to the percolation phenomena. When the insulation resistance of the suspending medium is not quite high, bit of electric current will flow and create additional resistive contact between conductive particles and then the mixture may be expected to show mild percolation phenomena.

In all cases, connectivity of dispersing particles strongly depend on the shape, properties of particle, so that the critical volume V_c should be adjusted depending on the properties of dispersing material, which relate to the geometrical parameters and the affinity between particles and orientation manner if the shape of powder has aspect ratio.

The validity of this equation is not only restricted to the permittivity case also it will be widely applicable to homogeneous and isotropic permeability problems, and also would be applicable to the resistive compound or colloidal dispersion of somewhat conductive powders and mediums.

3. Conclusions

The improved mixing rule equation has been refined and the application of this equation was expanded to the problems of dissipative or conductive powder and insulator mixture. The new equation would be useful for the material design of dielectric, magnetic or resistor compound and for the material design and evaluation of electromagnetic wave absorber.

More detailed consideration is needed with taking in account the physical, chemical, geometrical and some other parameters, which will relate to electromagnetic properties for very high frequency application.

Acknowledgement

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