

# Theory of dielectric relaxations due to the interfacial polarization for two-component suspensions of spheres

T. Hanai and K. Sekine

Institute for Chemical Research, Kyoto University, Uji, Kyoto, Japan

*Abstract:* A theoretical formula of dielectric relaxation in a form of complex relative permittivity is derived for dilute suspensions of spherical particles of two kinds on the basis of the Maxwell-Wagner theory of interfacial polarization. Another theoretical formula is derived further for concentrated suspensions of spheres of two kinds on condition that the formula derived above holds for the infinitesimally increasing process in concentration of the dispersed spheres. Furthermore a theoretical formula is derived for concentrated suspensions of shelled spheres of two kinds as the extension of the formula for concentrated suspensions. By use of the theoretical formulas proposed, values of the permittivities and the conductivities of the two-component suspensions were calculated for some examples with different sets of phase parameters. Results of the numerical calculation demonstrates dielectric relaxation profiles full of variety and characteristic of the suspensions containing two kinds of spheres covered with or without shells.

*Key words:* Dielectric relaxation, dielectric theory, interfacial polarization, relative permittivity, suspension.

## 1. Introduction

It is well known theoretically as well as experimentally that suspensions of particles in a continuous medium show dielectric relaxations due to interfacial polarization. Maxwell [1] and Wagner [2] proposed a dielectric theory of the interfacial polarization for a dilute suspension of spherical particles. Afterwards Hanai [3, 4] developed a dielectric theory of interfacial polarization for concentrated suspensions on the basis of the Maxwell-Wagner theory.

The dielectric relaxations predicted from the theories were discussed experimentally by many workers. The limiting values of the permittivities and conductivities at high and low frequencies in regard to the dielectric relaxations were discussed for a variety of emulsions [5]. The frequency dependence of the permittivities and the conductivities was also discussed in detail for W/O emulsions [6,7] and suspensions of ion exchange resin gel beads in water [8-11]. Furthermore the dielectric relaxations for the concentrated suspensions of spheres covered with a shell were formulated

and were successfully applied to the observations of polystyrene microcapsules [12, 13].

All the examples showed that the theory developed by Hanai for concentrated suspensions is in satisfactory agreement with the observed results as compared with the Maxwell-Wagner theory derived for dilute suspensions. At the present stage of the development of theories, it is desired to formulate and discuss the dielectric relaxation behavior of a concentrated suspension containing two kinds of dispersed particles; the suspension of this type is termed a two-component suspension hereinafter.

As regards dielectric theories for such two-component suspensions, Grosse [14] proposed an equation of the Bruggeman-Hanai type [15, 3] extended to two-component suspensions. Since conductivities are left out of theoretical consideration, dielectric relaxations due to the interfacial polarization cannot be discussed with his equation. Recently Boned and Peyrelasse [16] derived some theoretical formulas of complex permittivities for the case of multicomponent ellipsoidal

suspensions. Their discussion is of great use especially for such dilute ellipsoidal or spheroidal suspensions. Boyle [17] also derived theoretical equations for the permittivity and the conductivity of suspensions of an oriented dispersed phase of spheroidal shape applicable to higher concentrations. No attempt has so far been made to formulate the complex permittivity of two- or multicomponent suspensions.

In this paper, theoretical formulas for dilute two-component suspensions are first introduced on the basis of the Maxwell-Wagner theory. Next theoretical formulas are derived for concentrated two-component suspensions. Furthermore formulas are derived for concentrated two-component suspensions of shelled particles. Some frequency profiles are shown of permittivity and conductivity calculated from the theoretical formulas.

## 2. Extension of the Maxwell-Wagner theory to a dilute two-component suspension of spheres

Maxwell [1] and Wagner [2] presented a dielectric theory of interfacial polarization for a dilute suspension of spherical particles. Without loss of generality of the formulation, their theoretical formula can be extended to a suspension of dispersed particles of two kinds, henceforward termed  $j$ - and  $k$ -spheres, as the following:

$$\frac{\varepsilon^* - \varepsilon_a^*}{\varepsilon^* + 2\varepsilon_a^*} = \frac{\varepsilon_j^* - \varepsilon_a^*}{\varepsilon_j^* + 2\varepsilon_a^*} \Phi_j + \frac{\varepsilon_k^* - \varepsilon_a^*}{\varepsilon_k^* + 2\varepsilon_a^*} \Phi_k, \quad (1)$$

where  $\varepsilon^*$ ,  $\varepsilon_a^*$ ,  $\varepsilon_j^*$ , and  $\varepsilon_k^*$  denote the complex relative permittivity of the suspension, the continuous medium, the suspended  $j$ - and  $k$ -spheres, and  $\Phi_j$  and  $\Phi_k$  mean the volume fractions of the  $j$ - and  $k$ -spheres, respectively.

Asterisked permittivities  $\varepsilon^*$ 's are written as  $\varepsilon^* = \varepsilon + \kappa/(j\omega\epsilon_v)$  in terms of relative permittivity  $\varepsilon$ , electrical conductivity  $\kappa$ , angular frequency  $\omega$ , the permittivity of vacuum  $\epsilon_v$ , and imaginary unit  $j$ .

This Equation (1) is transformed to an explicit form with respect to  $\varepsilon^*$  as

$$\varepsilon^* = \varepsilon_a^* \frac{X}{Y}, \quad (2)$$

where

$$X = (\varepsilon_j^* + 2\varepsilon_a^*)(\varepsilon_k^* + 2\varepsilon_a^*) + 2(\varepsilon_k^* + 2\varepsilon_a^*)(\varepsilon_j^* - \varepsilon_a^*) \Phi_j + 2(\varepsilon_j^* + 2\varepsilon_a^*)(\varepsilon_k^* - \varepsilon_a^*) \Phi_k, \quad (2-X)$$

and

$$Y = (\varepsilon_j^* + 2\varepsilon_a^*)(\varepsilon_k^* + 2\varepsilon_a^*) - (\varepsilon_k^* + 2\varepsilon_a^*)(\varepsilon_j^* - \varepsilon_a^*) \Phi_j - (\varepsilon_j^* + 2\varepsilon_a^*)(\varepsilon_k^* - \varepsilon_a^*) \Phi_k. \quad (2-Y)$$

## 3. Derivation of the complex permittivity for a concentrated two-component suspension

### 3.1 Relation for infinitesimal increase of the dispersed phase in the continuous phase

It is assumed in the present theory that the dispersed phase to be added to the continuous phase is a mixture of the  $k$ - and the  $j$ -spheres with a fixed volume ratio  $K$ . At the final state in high concentrations, therefore, the volume fraction of the  $k$ -spheres  $\Phi_k$  and that of the  $j$ -spheres  $\Phi_j$  are related with each other by a relation

$$\frac{\Phi_k}{\Phi_j} = K. \quad (3)$$

The total volume fraction  $\Phi$  of the dispersed phase is given by

$$\Phi = \Phi_k + \Phi_j. \quad (4)$$

From Equations (3) and (4), we have

$$\Phi_j = \frac{1}{1+K} \Phi \quad \text{and} \quad \Phi_k = \frac{K}{1+K} \Phi. \quad (5)$$

Now we assume that the Maxwell-Wagner type Equation (1) holds for the infinitesimal increase in concentration such as that an infinitesimal quantity of the dispersed phase is added to the dispersion system of an arbitrary concentration. For such an infinitesimally increasing process of the dispersed phase,  $\varepsilon_a^*$ ,  $\varepsilon^*$  and  $\Phi$  in Equation (1) should be replaced as follows:

$$\varepsilon_a^* \rightarrow \varepsilon^*, \quad \varepsilon^* \rightarrow \varepsilon^* + \Delta\varepsilon^*, \quad \text{and} \quad \Phi \rightarrow \frac{\Delta\Phi'}{1-\Phi'}, \quad (6)$$

where  $\Delta\varepsilon^*$  denotes an increment of  $\varepsilon^*$  associated with this infinitesimally increasing process, and  $\Phi'$  means the volume fraction of the dispersed phase which is a mixture of the  $j$ - and the  $k$ -spheres. This replacement is the same as that adopted by Bruggeman [15] and Hanai [3] to derive dielectric formulas for concentrated suspensions.

Using Equations (5) and (6), a relation between  $\Delta\varepsilon^*$  and  $\Delta\Phi'$  on this infinitesimal process is derived from Equation (1) as

$$\frac{-\Delta\Phi'}{1-\Phi'} = \left[ \frac{\varepsilon^* - \varepsilon_j^*}{2\varepsilon^* + \varepsilon_j^*} + \frac{\varepsilon^* - \varepsilon_k^*}{2\varepsilon^* + \varepsilon_k^*} K \right]^{-1} \frac{1+K}{3\varepsilon^*} \Delta\varepsilon^*. \quad (7)$$

It is considered that a concentrated suspension is obtained as a result of a succession of these infinitesimally increasing processes. Mathematical representation of such a succession is the integration of Equation (7) over the ranges  $\Phi' [0, \Phi]$  and  $\varepsilon^* [\varepsilon_a^*, \varepsilon^*]$ .

### 3.2 Resolution into partial fractions of the infinitesimal relation

In order to perform the integral of the infinitesimal relation, Equation (7) must be rewritten in the form of partial fractions as follows:

$$\frac{-\Delta\Phi'}{1-\Phi'} = \frac{(2\varepsilon^* + \varepsilon_j^*)(2\varepsilon^* + \varepsilon_k^*) \Delta\varepsilon^*}{3\varepsilon^* [2(\varepsilon^*)^2 + \{(\varepsilon_j^* + \varepsilon_k^*) - 3(\varepsilon_j^* + \varepsilon_k^* K)/(1+K)\} \varepsilon^* - \varepsilon_j^* \varepsilon_k^*]}, \quad (8)$$

$$= \frac{(2\varepsilon^* + \varepsilon_j^*)(2\varepsilon^* + \varepsilon_k^*) \Delta\varepsilon^*}{3\varepsilon^* 2(\varepsilon^* - \alpha)(\varepsilon^* - \beta)}, \quad (9)$$

$$= \frac{2(\varepsilon^*)^2 + (\varepsilon_j^* + \varepsilon_k^*) \varepsilon^* + (1/2) \varepsilon_j^* \varepsilon_k^*}{3\varepsilon^* (\varepsilon^* - \alpha)(\varepsilon^* - \beta)} \Delta\varepsilon^*, \quad (10)$$

$$= \left( \frac{A}{\varepsilon^* - \alpha} + \frac{B}{\varepsilon^* - \beta} + \frac{C}{3\varepsilon^*} \right) \Delta\varepsilon^*. \quad (11)$$

The complex quantities  $\alpha$  and  $\beta$  in Equation (9) are the two roots of a quadratic in  $\varepsilon^*$  appearing in the denominator of Equation (8), being written as

$$\alpha = -\frac{1}{4} (M + \sqrt{M^2 + 8\varepsilon_j^* \varepsilon_k^*}), \quad (12)$$

$$\beta = -\frac{1}{4} (M - \sqrt{M^2 + 8\varepsilon_j^* \varepsilon_k^*}), \quad (13)$$

and

$$\alpha - \beta = -\frac{1}{2} \sqrt{M^2 + 8\varepsilon_j^* \varepsilon_k^*}, \quad (14)$$

where

$$M = -2(\alpha + \beta) = (\varepsilon_j^* + \varepsilon_k^*) - \frac{3(\varepsilon_j^* + \varepsilon_k^* K)}{1+K}. \quad (15)$$

After somewhat cumbersome calculation by comparison between Equations (10) and (11), the undetermined coefficients  $A$ ,  $B$  and  $C$  are determined as the following:

$$C = -1, \quad (16)$$

$$B = \frac{1}{2} - \frac{1}{4(\alpha - \beta)} \left[ (\varepsilon_j^* + \varepsilon_k^*) + \frac{\varepsilon_j^* + \varepsilon_k^* K}{1+K} \right], \quad (17)$$

and

$$A = \frac{1}{2} + \frac{1}{4(\alpha - \beta)} \left[ (\varepsilon_j^* + \varepsilon_k^*) + \frac{\varepsilon_j^* + \varepsilon_k^* K}{1+K} \right]. \quad (18)$$

### 3.3 Integration of the infinitesimal relation to derive the equation for concentrated suspensions

Integral of Equation (11) over the ranges  $\Phi' [0, \Phi]$  and  $\varepsilon^* [\varepsilon_a^*, \varepsilon^*]$  is written as

$$\int_0^\Phi \frac{-d\Phi'}{1-\Phi'} = \int_{\varepsilon_a^*}^{\varepsilon^*} \frac{1}{3\varepsilon^*} d\varepsilon^* + \int_{\varepsilon_a^*}^{\varepsilon^*} \frac{A}{\varepsilon^* - \alpha} d\varepsilon^* + \int_{\varepsilon_a^*}^{\varepsilon^*} \frac{B}{\varepsilon^* - \beta} d\varepsilon^*. \quad (19)$$

Complex integral calculation of Equation (19) leads to

$$\begin{aligned} \ln(1-\Phi) &= \frac{1}{3} [\text{Log } \varepsilon_a^* - \text{Log } \varepsilon^*] \\ &+ A[\text{Log}(\varepsilon^* - \alpha) - \text{Log}(\varepsilon_a^* - \alpha)] \\ &+ B[\text{Log}(\varepsilon^* - \beta) - \text{Log}(\varepsilon_a^* - \beta)], \quad (20) \end{aligned}$$

where  $\ln$  denotes the natural logarithm of real numbers, and  $\text{Log}$  means the principal value of the complex logarithm. This Equation (20) is the implicit function of  $\varepsilon^*$  for the concentrated two-component suspension

as a function of  $\varepsilon_j^*$ ,  $\varepsilon_k^*$ ,  $K$  and  $\Phi$  through Equations (12) to (15) and Equations (17) and (18).

It can readily be shown below that Equation (20) tends to the equation proposed formerly for a single component suspension. Under a condition  $\Phi_k = 0$  or  $K = 0$  which represents a single component suspension of the  $j$ -spheres only, one obtains  $M = \varepsilon_k^* - 2\varepsilon_j^*$ ,  $\alpha = -\varepsilon_k^*/2$ ,  $\beta = \varepsilon_j^*$ ,  $A = 0$ , and  $B = 1$  from Equations (12)–(18). Equation (20) is, therefore, simplified to the equation which was proposed in the previous paper [3, 4] for a single component suspension of  $j$ -spheres. For a suspension of  $k$ -spheres only, it follows that  $\Phi_j = 0$ ,  $K = \infty$ ,  $M = \varepsilon_j^* - 2\varepsilon_k^*$ ,  $\alpha = -\varepsilon_j^*/2$ ,  $\beta = \varepsilon_k^*$ ,  $A = 0$ , and  $B = 1$ . Equation (20) is simplified to the equation given in the previous papers for a  $k$ -sphere suspension. In the case of  $\varepsilon_k^* = \varepsilon_j^*$ , which means a single component suspension with the volume fraction  $\Phi$ , it turns out that  $M = -\varepsilon_j^*$ ,  $\alpha = -\varepsilon_j^*/2$ ,  $\beta = \varepsilon_j^*$ ,  $A = 0$ , and  $B = 1$ . Equation (20) is also simplified to the equation for a  $j$ -sphere suspension.

#### 4. Extension of the equation of concentrated suspension to a two-component suspension of shelled spheres

From a dielectric point of view, several examples such as microcapsules, lipid vesicle suspensions and biological cell suspensions are considered to be suspensions of shelled spheres.

Dielectric formulation for this kind of concentrated suspension of shelled spheres can be achieved by introducing equivalent complex relative permittivities  $\varepsilon_{qj}^*$  and  $\varepsilon_{qk}^*$  of the shelled  $j$ -spheres and  $k$ -spheres in the same manner as used previously in the dielectric analysis of microcapsules [12, 13], lipid vesicles [18] and erythrocyte suspensions [19].

The formula of  $\varepsilon_{qj}^*$  for the  $j$ -spheres covered with the  $j$ -shells and that of  $\varepsilon_{qk}^*$  for the  $k$ -spheres covered with the  $k$ -shells are given as follows:

$$\varepsilon_{qj}^* = \varepsilon_{sj}^* \frac{2(1 - v_j) \varepsilon_{sj}^* + (1 + 2v_j) \varepsilon_{ij}^*}{(2 + v_j) \varepsilon_{sj}^* + (1 - v_j) \varepsilon_{ij}^*}, \quad (21)$$

$$\varepsilon_{qk}^* = \varepsilon_{sk}^* \frac{2(1 - v_k) \varepsilon_{sk}^* + (1 + 2v_k) \varepsilon_{ik}^*}{(2 + v_k) \varepsilon_{sk}^* + (1 - v_k) \varepsilon_{ik}^*}, \quad (22)$$

$$v_j = \left(1 - 2 \frac{d_j}{D_j}\right)^3, \quad (23)$$

and

$$v_k = \left(1 - 2 \frac{d_k}{D_k}\right)^3, \quad (24)$$

where  $d$  and  $D$  are the thickness and the outer diameter of the spherical shell,  $\varepsilon_s^*$  and  $\varepsilon_i^*$  are the complex relative permittivity of the shell phase and of the inner phase, and the subscripts  $j$  and  $k$  refer to the  $j$ -spheres and the  $k$ -spheres, respectively.

For this concentrated suspension of shelled spheres, the quantities  $\alpha$ ,  $\beta$ ,  $\alpha - \beta$ ,  $M$ ,  $A$  and  $B$  are expressed by the following formulas instead of by Equations (12) to (15) and Equations (17) and (18):

$$\alpha = -\frac{1}{4} (M + \sqrt{M^2 + 8\varepsilon_{qj}^* \varepsilon_{qk}^*}), \quad (25)$$

$$\beta = -\frac{1}{4} (M - \sqrt{M^2 + 8\varepsilon_{qj}^* \varepsilon_{qk}^*}), \quad (26)$$

$$\alpha - \beta = -\frac{1}{2} \sqrt{M^2 + 8\varepsilon_{qj}^* \varepsilon_{qk}^*}, \quad (27)$$

$$M = (\varepsilon_{qj}^* + \varepsilon_{qk}^*) - \frac{3(\varepsilon_{qj}^* + \varepsilon_{qk}^* K)}{1 + K}, \quad (28)$$

$$A = \frac{1}{2} + \frac{1}{4(\alpha - \beta)} \left[ (\varepsilon_{qj}^* + \varepsilon_{qk}^*) + \frac{\varepsilon_{qj}^* + \varepsilon_{qk}^* K}{1 + K} \right], \quad (29)$$

and

$$B = \frac{1}{2} - \frac{1}{4(\alpha - \beta)} \left[ (\varepsilon_{qj}^* + \varepsilon_{qk}^*) + \frac{\varepsilon_{qj}^* + \varepsilon_{qk}^* K}{1 + K} \right]. \quad (30)$$

Consequently the complex relative permittivity  $\varepsilon^*$  of the two-component suspension of shelled spheres is given by Equation (20) as an implicit function of  $\varepsilon_{sj}^*$ ,  $\varepsilon_{sk}^*$ ,  $\varepsilon_{ij}^*$ ,  $\varepsilon_{ik}^*$ ,  $d_j$ ,  $d_k$ ,  $D_j$ ,  $D_k$ ,  $K$  and  $\Phi$  through Equations (21) to (30).

#### 5. Some numerical examples of the dielectric relaxation

In order to examine the frequency profile of the permittivity and the conductivity of suspensions of these kinds, numerical calculation was carried out for some examples with different sets of phase parameters by means of the preceding formulas. Four typical examples, termed Systems,  $A$ ,  $B$ ,  $C$  and  $D$ , are specified by each set of phase parameters, whose values are listed in Table 1.

Table 1. Values of phase parameters characterizing Systems A, B, C and D for evaluating theoretical curves of the dielectric relaxation due to the interfacial polarization

	System A	System B	System C	System D
Constitution	concentrated two-component suspensions without shells	concentrated two-component suspensions of shelled spheres		
Equations concerned	Equations (12) to (20)	Equations (20) to (30)		
Continuous medium:				
$\epsilon_a$	3	80	80	80
$\kappa_a/\mu\text{S cm}^{-1}$	0	1	1	1
Shell phase of the dispersed particles: $D_j = D_k = 500 \mu\text{m}$ , $d_j = d_k = 2 \mu\text{m}$				
$j$ -shell, $\epsilon_{sj}$	—	3	3	3
$\kappa_{sj}/\mu\text{S cm}^{-1}$	—	0	0	1
$k$ -shell, $\epsilon_{sk}$	—	3	3	3
$\kappa_{sk}/\mu\text{S cm}^{-1}$	—	0	0	0
Inner phase of the dispersed particles:				
$j$ -sphere, $\epsilon_{ij}$	80	80	80	80
$\kappa_{ij}/\mu\text{S cm}^{-1}$	1	1	30	$10^3$
$k$ -sphere, $\epsilon_{ik}$	80	80	80	80
$\kappa_{ik}/\mu\text{S cm}^{-1}$	$10^3$	$10^3$	$10^3$	$10^5$
Volume fraction of the dispersed particles:				
total, $\Phi$	0.8	0.6	0.6	0.6
$j$ -sphere, $\Phi_j$	0.8–0	0.6–0	0.6–0	0.6–0
$k$ -sphere, $\Phi_k$	0–0.8	0–0.6	0–0.6	0–0.6
Number of dielectric relaxations	2	2	3	4
Figures concerned	Figures 1, 2	Figures 3, 4	Figures 5, 6	Figures 7, 8

### 5.1 Two-component suspension without shells

A suspension, termed System A, is assumed to be composed of two kinds of dispersed particles with respect to the conductivity of inner phases. The relative permittivity  $\epsilon$  and the conductivity  $\kappa$  of System A at each frequency can be calculated by means of Equations (3)–(5), (12)–(18) and (20) for different values of the concentration  $\Phi_j$  under a fixed value  $\Phi = 0.8$ . The theoretical curves of the dielectric relaxation obtained are shown in Figure 1, in which two distinct dielectric relaxations are found for the mixed systems of  $j$ - and  $k$ -spheres.

Figure 2 shows the complex plane plots of the complex relative permittivity for a case, termed System A-d, with  $\Phi_j = 0.1$  in System A. Two circular arc loci can readily be found in the figure, being indications of two dielectric relaxations.

### 5.2 Two-component suspension of shelled spheres

The suspensions, termed System B, C and D in Table 1, are all assumed to contain two kinds of shelled spheres in such a manner that the inner phase conductivity of  $j$ -spheres is different from that of  $k$ -spheres: namely  $\kappa_{ij} \neq \kappa_{ik}$ . The outer diameter and the shell thickness of the shelled spheres are assumed as  $D_j = D_k = 500 \mu\text{m}$  and  $d_j = d_k = 2 \mu\text{m}$ . The relative permittivity  $\epsilon$  and the conductivity  $\kappa$  of these Systems at each frequency can be calculated by means of Equations (3)–(5) and (20)–(30) for different values of the concentration  $\Phi_j$  under a fixed value  $\Phi = 0.6$ .

In System B, the inner phase conductivity of  $j$ -spheres  $\kappa_{ij}$  is assumed to be equal to that of the continuous medium  $\kappa_a$ . The frequency dependence of  $\epsilon$ ,  $\kappa$  and loss factor  $\Delta\epsilon''$  obtained is shown in Figure 3. The data of complex relative permittivity for a case, termed

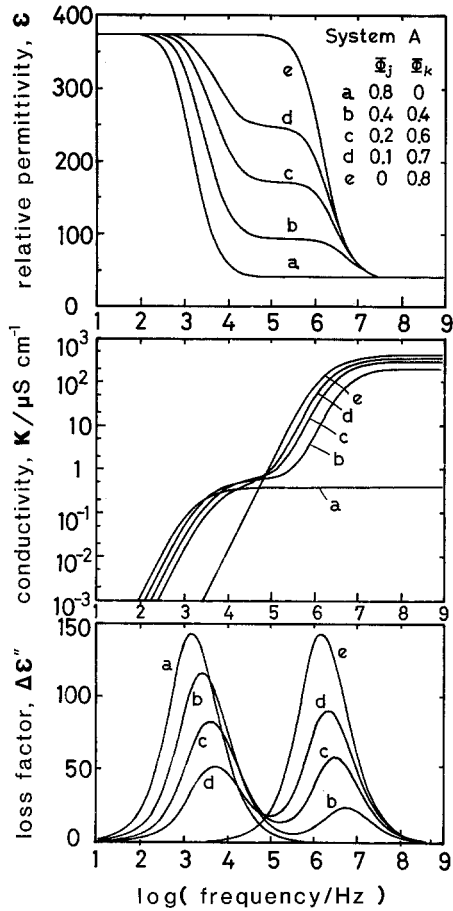


Fig. 1. Frequency dependence of relative permittivity  $\epsilon$ , electrical conductivity  $\kappa$  and loss factor  $\Delta\epsilon'' = (\kappa - \kappa_i)/(2\pi f \epsilon_v)$  for System A. The curves are calculated by means of Equations (3)–(5), (12)–(18) and (20) with the values of phase parameters listed in Table 1

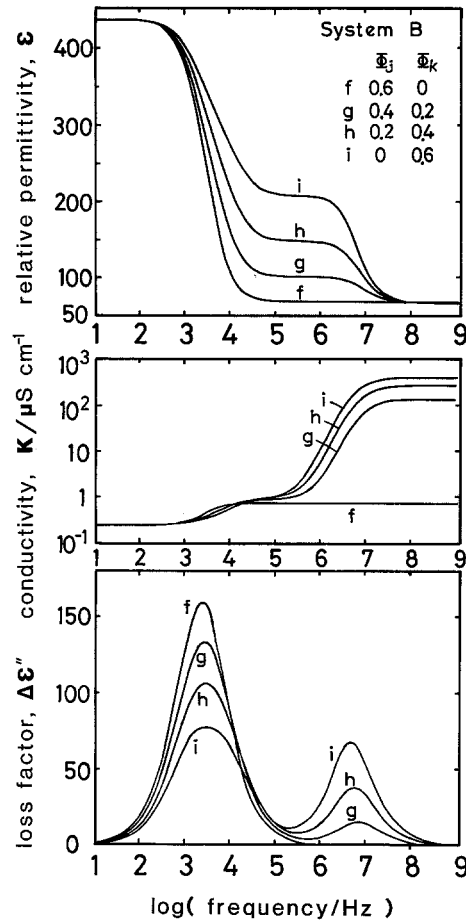


Fig. 3. Frequency dependence of  $\epsilon$ ,  $\kappa$  and  $\Delta\epsilon'' = (\kappa - \kappa_i)/(2\pi f \epsilon_v)$  for System B. The curves are calculated by means of Equations (3)–(5), (20)–(30) with the values of phase parameters listed in Table 1

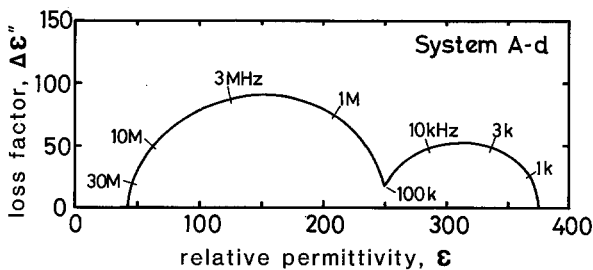


Fig. 2. Complex plane plots of relative permittivity  $\epsilon$  and loss factor  $\Delta\epsilon''$  for System A-d with  $\Phi_j = 0.1$ . Numbers along the curve are the frequency. The values of phase parameters used are shown in Table 1

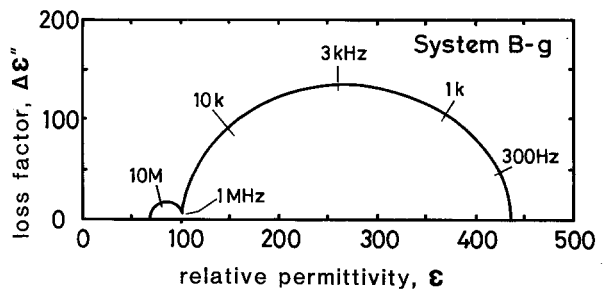


Fig. 4. Complex plane plots of  $\epsilon$  and  $\Delta\epsilon''$  for System B-g with  $\Phi_j = 0.4$ . Numbers along the curve are the frequency

System B-g, with  $\Phi_j = 0.4$  in System B are plotted in Figure 4. As readily seen in Figures 3 and 4, System B shows two distinct dielectric relaxations.

In System C, the inner phase conductivity of  $j$ -spheres  $\kappa_{ij}$  is assumed to be different from that of the continuous medium  $\kappa_a$ . The frequency dependence of

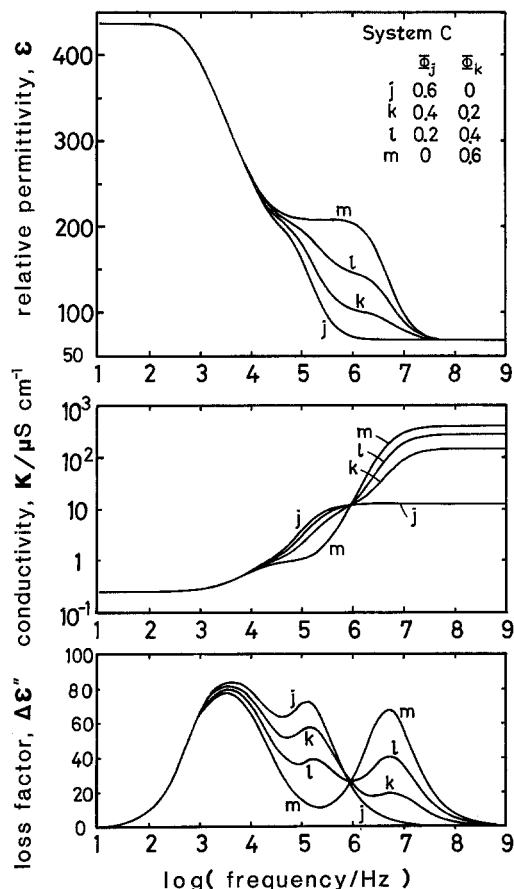


Fig. 5. Frequency dependence of  $\epsilon$ ,  $\kappa$  and  $\Delta\epsilon'' = (\kappa - \kappa_i)/(2\pi f \epsilon_v)$  for System C. The values of phase parameters used are listed in Table 1

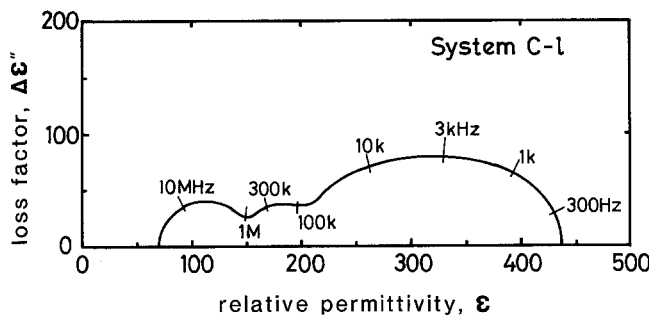


Fig. 6. Complex plane plots of  $\epsilon$  and  $\Delta\epsilon''$  for System C-l with  $\Phi_j = 0.2$

$\epsilon$ ,  $\kappa$  and loss factor  $\Delta\epsilon''$  calculated for System C is shown in Figure 5. The data, termed System C-l, for  $\Phi_j = 0.2$  in System C are plotted on the complex relative permittivity plane in Figure 6. As readily seen in Figures 5 and 6, System C shows three dielectric relaxations.

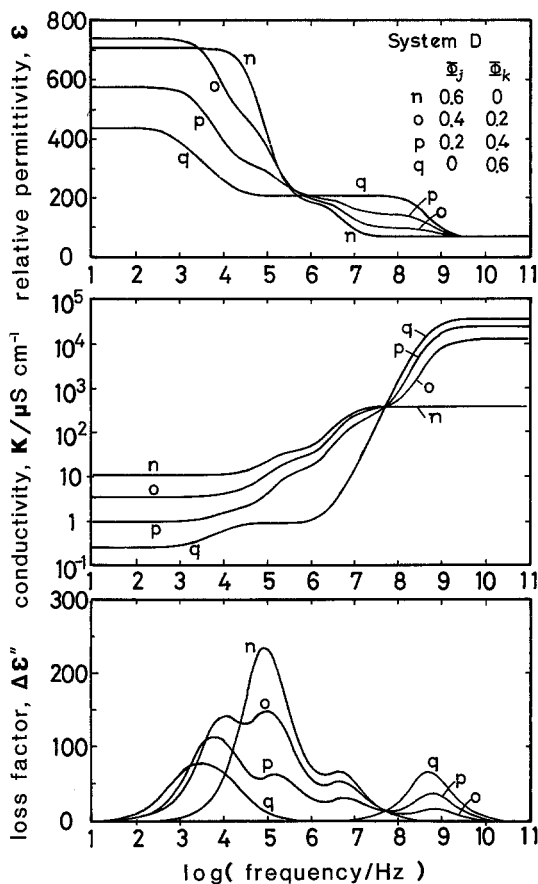


Fig. 7. Frequency dependence of  $\epsilon$ ,  $\kappa$  and  $\Delta\epsilon'' = (\kappa - \kappa_i)/(2\pi f \epsilon_v)$  for System D. The values of phase parameters used are listed in Table 1

A suspension, termed System D, is characterized by a set of phase parameters as shown in Table 1 with  $\kappa_{sj} = 1 \mu\text{S cm}^{-1}$  in addition to  $\kappa_{ij} = 10^3 \mu\text{S cm}^{-1}$ . The frequency dependence of the relative permittivity  $\epsilon$ , the conductivity  $\kappa$  and the loss factor  $\Delta\epsilon''$  calculated for System D is shown in Figure 7. The data, termed System D-p, for  $\Phi_j = 0.2$  in System D are plotted on the complex relative permittivity plane in Figure 8. This System D is seen in Figures 7 and 8 to show four dielectric relaxations.

It is concluded that Equation (20) gives different frequency profiles of permittivities and conductivities for concentrated two-component suspensions of particles covered with or without shells. The theory and the equations proposed in the present work can be applied to dielectric relaxation data on emulsions, suspensions, microcapsules, lipid vesicle suspensions and biological cell suspensions to obtain more valuable information concerning the inner structure of these disperse systems.

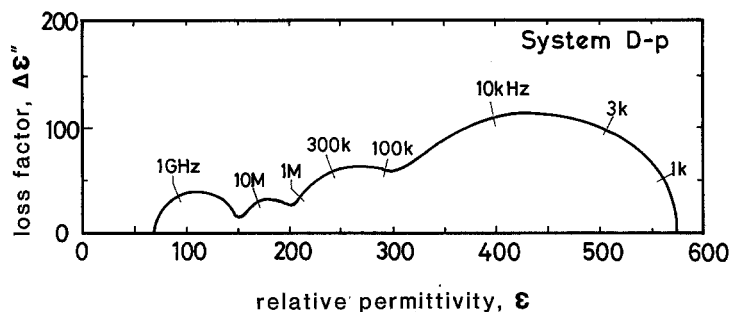


Fig. 8. Complex plane plots of  $\epsilon$  and  $\Delta\epsilon''$  for System D-p with  $\phi_j = 0.2$

### References

- Maxwell JC (1891) *A Treatise on Electricity and Magnetism*, Third ed, Vol 1, Chap 9, Art 310–314, Clarendon Press, Oxford, p 435
- Wagner KW (1914) *Arch Electrotech* 2:371
- Hanai T (1960) *Kolloid Z* 171:23
- Hanai T (1961) *Kolloid Z* 175:61
- Hanai T (1968) In: Sherman P (ed) *Electrical Properties of Emulsions*, *Emulsions Science*, Chap 5, Academic Press, London, New York, p 353
- Hanai T (1961) *Kolloid Z* 177:57
- Hanai T, Imakita T, Koizumi N (1982) *Coll & Polym Sci* 260:1029
- Ishikawa A, Hanai T, Koizumi N (1981) *Jpn J Appl Phys* 20:79
- Ishikawa A, Hanai T, Koizumi N (1982) *Jpn J Appl Phys* 21:1762
- Ishikawa A, Hanai T, Koizumi N (1983) *Jpn J Appl Phys* 22:942
- Ishikawa A, Hanai T, Koizumi N (1984) *Coll & Polym Sci* 262:477
- Zhang HZ, Sekine K, Hanai T, Koizumi N (1983) *Coll & Polym Sci* 261:381
- Zhang HZ, Sekine K, Hanai T, Koizumi N (1984) *Coll & Polym Sci* 262:513
- Grosse C (1979) *J Chim Phys* 76:153
- Bruggeman DAG (1935) *Ann Phys Leipzig* (5) 24:636
- Boned C, Peyrelasse J (1983) *Coll & Polym Sci* 261:600
- Boyle MH (1985) *Coll & Polym Sci* 263:51
- Sekine K, Hanai T, Koizumi N (1983) *Bull Inst Chem Res Kyoto Univ* 61:299
- Hanai T, Asami K, Koizumi N (1979) *Bull Inst Chem Res Kyoto Univ* 57:297

Received March 27, 1986;  
accepted April 22, 1986

### Authors' address:

Tetsuya Hanai and Katsuhisa Sekine  
Institute for Chemical Research  
Kyoto University  
Uji, Kyoto 611, Japan