

The Material Determination of Promoting ER Effects Using Core-Shell or Single Solid Phase Particulates Suspended in an Electrorheological Fluid

I-Chung Cheng^{*}, Woei-Shyong Lee^{**}, Chang Chuen Sun^{***},
and Wen-Hwa Hwu^{*}

^{*}Department of Applied Chemistry, Chung Cheng Institute of Technology, National Defense University

^{**}Mackay Medicine, Nursing and Management College

^{***}Department of Electronic Engineering, Lee-Ming Institute of Technology

ABSTRACT

The influences of the structure and the material of suspended particles on electrorheological effects have been investigated. The suspended particles with a core-shell structure or a single solid phase structure were immersed in an electrorheological fluid and were applied by an external field. Based on the dielectric polarized model, the electric potential which satisfies Laplace's equation was solved by using the point-dipole approximation method. The results have shown that the inter-particle forces in electrorheological fluids are apparently enhanced after using the core-shell structural suspended particles rather than using single phase solid particles. However, the replacing material or coating material must have a higher dielectric constant than that of the original particles either changing the core material of core-shell particles or coating a shell material on the single phase solid particles, to promote the ER effects.

Keywords: Suspended particle, Electrorheological Effect, Dielectric polarization model, Core-shell structure

提升核殼或實心結構粒子電流變效應之材料選擇

鄭義忠^{*} 李偉雄^{**} 孫長春^{***} 胡文華^{*}

^{*}國防大學中正理工學院應用化學研究所

^{**}馬偕醫護管理專科學校

^{***}黎明技術學院電子工程系

摘 要

本文針對具複合核殼結構或單一實心結構之懸浮固體粒子，浸於電流變液中並受到電場作用下，討論選擇結構與材料對電流變效應的影響。分析過程係根據介電極化模型理論，採用點偶極方法分析求解拉普拉斯方程式獲得電位分佈。由計算結果知，利用核殼的方法可提昇原有單一材料實心粒子的作用力，然而不論是使用核心改質或外層披覆，為了提升電流變效應，應選用較原材料更高介電係數的材料。

關鍵詞：懸浮粒子，電流變效應，介電極化模型，核殼結構

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I. INTRODUCTION

The electrorheological fluid (ERF), a novel and smart material among the new materials, was invented during the twentieth century. The ERF's apparent viscosity change is abruptly acted under an electric field and this change is called as the electrorheological effect, discovered by Winslow in 1949 [1]. An ERF is a suspension with dielectric solid particles in an insulating host liquid. Its apparent viscosity is higher under an external field than without an applied field and it is nearly solidified. The yield stress commonly appeared in solids can be observed in this solid-like fluid.

Since the electrorheological characteristics of an ERF, apparent viscosity and yield stress, can be controlled by an external field, the ERF's quick response to an action is within milliseconds and has been widely used in various fields of mechanical applications, such as actuators, vibration suppressions and isolations, hydraulic valves, robotics, and intelligent action mechanism [2]. Its excellent mechanical controllability can largely reduce the device complexity and cost, improves the control performance of the system, and accomplishes the functions which conventional mechanical devices are hardly to achieve. Since the ERF has a great potential in applications, such as aeronautics and aviation, production automation, armament control, robotic engineering, noise control, automobile engineering, watercraft engineering, hydraulic engineering, agriculture machinery and physical exercising apparatus, the ER techniques are broadly researched in the past half century [3].

In order to promote the yield stress applied by the external field, a variety of materials, such as inorganic, semi-conducting organic, composite material, have been selected to make dispersed particles to form an ER fluid [4-14]. Among them, the composite material is a main subject in the recent studies. The ER devices, such as clutches, brakes, actuators, valves and damping devices, are still in the developing stage for the commercial purpose after a half century. The main reason why the ER fluid is still not widely used is that the yield stress is not high enough and that the ER system is not wholly stable yet [5].

In general, there are two classifications of ER fluids: wet-base system and dry-base system.

The wet-base system was the main issue in the early studies and the solid particles used were largely inorganic material or nature substances [4]. In wet-base system, the additives, such as water or surfactants, were added to activate the ER system to improve the ER effects significantly. However, the wet-base ER system containing small molecular additives has temperature limitations in practical applications. It will badly affect the ER effects that the small molecular additives will vaporize at higher temperature while they will condense at lower temperature. In order to avoid the disadvantages of the wet-base system, the dry-base system were selected to work on the ER fluid. The particles chosen for the dry-base system were typically semi-conductive and intrinsically polarizable solid particles. The dry-base ER system has a wide range of working temperature because of the lack of additives. The particles commonly used are aluminosilicate, carbonaceous, semi-conducting polymers, organic-composite particles [4-14].

In theoretical investigations of an ER fluid, after the dry-base system was proposed, the researchers were forced to give up the electric double layer (EDL) model which was well explained the observation in the wet-base system [4, 15] and started to accept the electric properties mismatch model [4, 8, 16]. In the EDL model, the additives are significant and necessary in order to reveal an ER effect, however, the dry-base ER system can show an obvious ER response to an action without the existence of additives. For dry-base systems, the theoretical models proposed by researchers are polarization model, conduction model and dielectric loss model [4]. No matter which model was adopted, a concept largely accepted is that the abrupt change of ER properties is strongly related to the structure

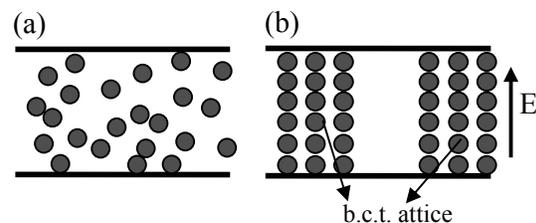


Fig. 1. Schematic illustration of the structure change of an ERF before (a) and after (b) an external electric field is applied.

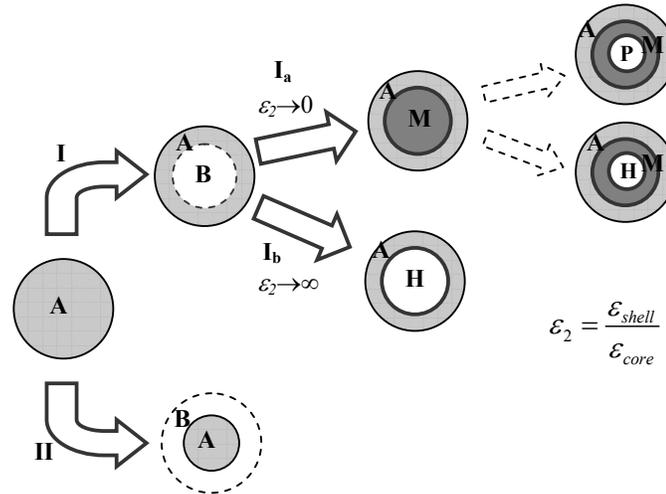


Fig. 2. ER particles enhancement schemes. A single phase solid particle is changed to a core-shell structural composite particle (Route I). Followed by either replacing the centered core by a metal ($\epsilon_2 \rightarrow 0$; Route I_a) or keeping the centered core empty to form a hollow sphere ($\epsilon_2 \rightarrow \infty$; Route I_b). Route II: Coating a shell on the single phase solid particle to form a core-shell particle. (A: Original particle material; B: New material except for A; M: Metal; H: Hollow; P: Polymer).

changes under the influence of an applied field. The strong inter-particle forces result in the high yield stress. A typical microstructure change of an ER fluid, before and after an electric field is applied, is shown in Figure 1. Before an electric field is applied, the particles in a dielectric fluid are randomly distributed, while after an external field is applied the dispersed particulates oriented along the direction of the applied field. The microstructure change from the disordered state to an ordered state is that the dispersed particulates produce dipoles induced by the external field and the dispersed particles are attracted together acted by the dipole-dipole interaction force. The attracted particles are oriented to form particle chains along the direction of the applied electric field and finally form stable particle columns. Within a column the particles are arrayed as a crystallite of a body-centered tetragonal (bct) lattice, which can be determined with a laser diffraction technique [17].

In the recent years, there were many works published about the dispersed particles made of composite materials other than the single phase solid material so as to promote the ER effects [18]. But, the related theoretical analyses are mostly based on the numerical computations. The experimental observation is hardly to be

seen. Therefore, we focused our goal on the core-shell structural particles in this paper. The dielectric polarized model was utilized for the composite particles with a core-shell structure to derive an analytic expression of the inter-force between the particles by means of a dipole approximation. Thereafter, the solution to the core-shell structural case is compared with the solution to the single phase solid particle case. The structures are determined and the materials are chosen theoretically so that the ER particles with the core-shell structure have higher performances than that with a single solid phase. The predicted results will be the basis of fabricating the ER particles in the future work. The schematic diagram of a single phase solid particle material improvement is shown in Figure 2. A single phase solid particle is changed to a core-shell structural composite particle (Route I). It is followed by either replacing the centered core by a metal ($\epsilon_2 \rightarrow 0$; Route I_a) or keeping the centered core empty to form a hollow sphere ($\epsilon_2 \rightarrow \infty$; Route I_b). The other scheme (Route II) is to coating a shell on the single phase solid particle to form a core-shell particle.

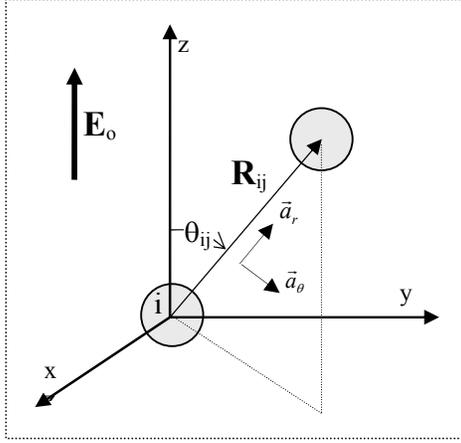


Fig 3. Coordinate system used to define the pair interaction due to electrical polarization [Eq. 1]. E_o is the external electric field.

II. MATHEMATICAL MODEL

2.1 Single Phase Solid Particles

The interforce between two single solid phase ER particles can be expressed by the dipole approximation [4, 19, 20]. The coordinate system used to define the pair interaction due to electric polarization is shown in Figure 3. The inter-particle force is given in Equation 1.

$$\vec{F}_{ij}(r, \theta) = \frac{3p^2}{4\pi\epsilon_f r^4} [(3\cos^2\theta - 1)\vec{a}_r + (\sin 2\theta)\vec{a}_\theta] \quad (1)$$

where

$$p = 4\pi\epsilon_f \beta b^3 E_o \quad (2)$$

$$\beta = \frac{\alpha - 1}{\alpha + 2}; \quad \alpha = \frac{\epsilon_p}{\epsilon_f} \quad (3a,b)$$

and p is the dipole moment, b the particle radius, ϵ_f the permittivity of the insulating fluid, ϵ_p the permittivity of the particle, E_o the external electric field, r the distance between two particle centers, \vec{a}_r , \vec{a}_θ the spherical coordinate unit electric field, r the distance between two particle vectors in the r and θ directions, respectively.

Equation 1 is only suitable for the case of two polarized particles separated far enough. Once two polarized particles are very close, they will affect each other. Ultimately, they will be polarized much further. The polarizations of both particles are increased and the interforce between two particles is increased. Considering the effect of the multipole moment, the total

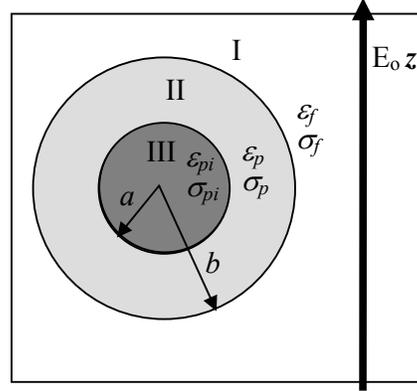


Fig. 4. Schematic drawing of a particle with a core-shell structure. The thickness of the shell is $b-a$, where b is the radius of the particle and a is the radius of the inner core (I: oil zone, II: shell zone, III: core zone).

dipole moment can be expressed as follows [21].

$$p_r = p \sum_{n=0}^{\infty} (-\beta)^n \left(\frac{\sinh\left(\frac{r}{2b}\right)}{\sinh\left[(n+1)\frac{r}{2b}\right]} \right)^3 \quad (4)$$

Equation 4 is obtained from multiplying a correction term, a value larger than one, on Equation 2. From Equation 1, 2 and 3, the inter-particle force will increase with p^2 increasing, that is, the interforce will increase with β^2 increasing. If the particle material is correctly determined to form a core-shell structural particle, high yield stresses of an ERF will be reached.

2.2 Core-Shell Particle

Considering a particle with a core-shell structure and with a radius of b , its core has a radius of a and a permittivity of ϵ_{pi} . The shell portion of the particle has a thickness of $b-a$ and a permittivity of ϵ_p . The core-shell particle is immersed in an insulating host liquid with a permittivity of ϵ_f and a uniform external electric field, E_o , is applied. The schematic drawing of the particle with a core-shell structure is shown in Figure 4.

The electric potential satisfies Laplace's equation $\nabla^2 V = 0$ on insulating liquid zone (zone I), shell zone (zone II), and core zone (zone III), respectively. D_n denotes the normal component of the dielectric displacement (\mathbf{D}).

The potentials at the surface and the interface of the sphere are continuous so that there are six boundary conditions

$$\begin{aligned} r \rightarrow \infty \quad V^I &= -E_o r \cos \theta \\ r = b \quad V^I &= V^{II}, D_n^I = D_n^{II} \\ r = a \quad V^{III} &= V^{II}, D_n^{III} = D_n^{II} \\ r = 0 \quad V^{III} &= \text{finite} \end{aligned} \quad (5)$$

In Equation 5, the potential and the normal component of \mathbf{D} are continuous across the surface ($r = b$) and the interface ($r = a$). The potential of the particle in zone I is solved through the Laplace's equation associated with boundary conditions and it is expressed as a general form, Equation 6.

$$V^I(r, \theta) = -\left(r - \frac{\beta^* b^3}{r^2}\right) E_o \cos \theta ; r \geq b \quad (6a)$$

$$V^{II}(r, \theta) = -f^* \left(r + \frac{g^*}{r^2}\right) E_o \cos \theta ; a \leq r \leq b \quad (6b)$$

$$V^{III}(r, \theta) = -h^* r E_o \cos \theta ; r \leq a \quad (6c)$$

where

$$\beta^* = \frac{b^3(2\varepsilon_2 + 1)(\varepsilon_1 - 1) - a^3(\varepsilon_2 - 1)(2\varepsilon_1 + 1)}{b^3(\varepsilon_1 + 2)(2\varepsilon_2 + 1) - 2a^3(\varepsilon_1 - 1)(\varepsilon_2 - 1)} \quad (7)$$

$$f^* = \frac{3(2\varepsilon_2 + 1)b^3}{b^3(\varepsilon_1 + 2)(2\varepsilon_2 + 1) - 2a^3(\varepsilon_1 - 1)(\varepsilon_2 - 1)} \quad (8a)$$

$$g^* = \frac{\varepsilon_2 - 1}{2\varepsilon_2 + 1} a^3 \quad (8b)$$

$$h^* = \frac{9\varepsilon_2 b^3}{b^3(\varepsilon_1 + 2)(2\varepsilon_2 + 1) - 2a^3(\varepsilon_1 - 1)(\varepsilon_2 - 1)} \quad (8c)$$

$$\varepsilon_1 = \frac{\varepsilon_p}{\varepsilon_f} \quad (8d)$$

$$\varepsilon_2 = \frac{\varepsilon_p}{\varepsilon_{pi}} \quad (8e)$$

By the dipole approximation and from the expression of the potential in zone I, the effective dipole moment of the core-shell structural composite particles is concluded to be Equation 8.

$$p = 4\pi\varepsilon_f \beta^* b^3 E_o \quad (9)$$

Comparing Equation 9 to Equation 2, β is simply replaced by β^* . For the convenience, Equation 7 is rearranged as

$$\beta^* = \frac{\alpha^* - 1}{\alpha^* + 2} \quad (10a)$$

where

$$\alpha^* = \left(\frac{\varepsilon_p}{\varepsilon_f}\right) \left(\frac{M-2}{M+1}\right) \quad (10b)$$

$$M = \frac{b^3(2\varepsilon_2 + 1)}{a^3(\varepsilon_2 - 1)} \quad (10c)$$

α^* is the dielectric mismatch ratio of the core-shell particle to the host liquid.

In Equation 10, when ε_2 is equal to one, $\varepsilon_2=1$, the permittivity of the core and shell materials is the same. Thus, the α^* in Equation 10b can be simplified to Equation 3b. Comparing Equation 3b to 10b, for the case of $\varepsilon_2 \neq 1$, the α^* in the core-shell structural formula has one more factor, $(M-2)/(M+1)$, than the α in the single phase solid particle expression. This factor is a function of the particle structure, the permittivity ratio of the shell material to the core material.

III. RESULTS AND DISCUSSION

Figure 5 illustrates the effect of force, represented by $(\beta^*)^2$, versus the dielectric mismatch ratio, α^* . There exists inter-particle force, that is $(\beta^*)^2 \neq 0$, except for $\alpha^*=1$. The dielectric mismatch of two materials (particle and liquid) results in the strong ER effect [4, 19, 20]. This is always emphasized by the conventional polarization model.

The special behavior of an ERF within the region of $\alpha^* < 1$ expressed by the dashed line in Figure 5 is due to the inverse polarization. The inverse polarization happens when the direction of the dipole induced on particles by an external electric field is opposite to the direction of the applied electric field. The particles immersed in an ERF will rotate constantly acted by torsions. This model has been proposed by Boissy and his colleagues [22]. The particle chains are unstable and eventually the chains will collapse. The collapsed particles will shift to electrode plates by the electrophoresis effect and the liquid-solid separation will happen. As a result, the yield stress decreases as the external electric field increases. This is termed the negative ER effect. In Figure 5, the values on the dashed line within the region of $\alpha^* < 1$ indicate the maximum magnitude of instantaneous attractive forces rather than the yield stresses of an ERF due to the unstable particles chains exist. However, for the region of $\alpha^* > 1$ expressed by a solid line, the particle chains are stable and thus the attractions between particles are strong, the shear stresses

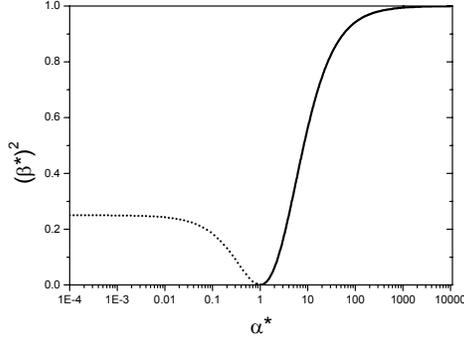


Fig. 5. $(\beta^*)^2$ as a function of α^* for a core-shell structural particle. The dashed line expresses the unstable polarization region.

are high, and the yield stresses are large. From Figure 5, the $(\beta^*)^2$ increases with α^* increasing. Traditionally, in order to have a higher yield stress, the ER system always contains the single phase solid particles with high dielectric constants. These particles are dispersed in an insulating liquid with a very low dielectric constant [4].

Following discussions are two schemes we have tried to promote the ER particles performance. (1) Changing a single phase solid particle into a core-shell structural composite particle. (2) Coating a shell on the single phase solid particle to form a core-shell particle.

Scheme I: Changing the core material of a core-shell particle

The first scheme is changing a single phase solid particle made of material A into a core-shell structural composite particle as shown in route I of Figure 2. The composite particle has material B in the centered core and enclosed by a material A as a shell. Following the route I, routes I_a and I_b depict two efforts that the centered core with a material B is either replaced by a metal, $\varepsilon_2 \rightarrow 0$, or is kept empty to form a hollow sphere, $\varepsilon_2 \rightarrow \infty$.

For the case of the single phase solid particle, $\alpha = \varepsilon_p / \varepsilon_f$ expresses the dielectric mismatch while $\alpha^* = (\varepsilon_p / \varepsilon_f) \{ (M-2) / (M+1) \}$ expresses the dielectric mismatch for the core-shell particle case. As long as $\varepsilon_2 \neq 1$, the α^* in the core-shell structure has one more factor, $(M-$

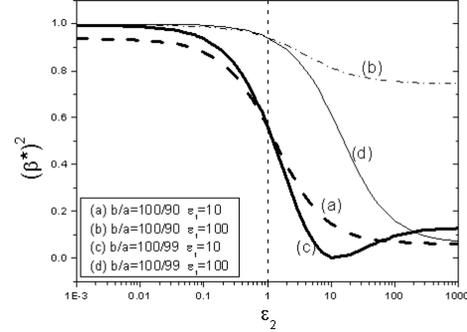


Fig. 6. $(\beta^*)^2$ as a function of ε_2 for a core-shell structural particle at different geometrical and electric properties conditions.

$2) / (M+1)$, than the α in the single phase case. This factor is a function of the particle structure and the ratio of the permittivity of shell material to the permittivity of core material.

In general, the shell portion of a core-shell structural particle is not too thick, thus, the ratios of b/a are set as 100/90 and 100/99. The relations of $(\beta^*)^2$ versus ε_2 for particles with a core-shell structure were computed using Equations 10. The results are shown in Figure 6 for different dielectric ratios, $\varepsilon_i = 10$ and 100, under the conditions of $b/a = 100/90$ and 100/99. The values of $(\beta^*)^2$ corresponding to the single phase solid particles are the intersection points between curves and the dashed vertical line, $\varepsilon_2 = 1$, in Figure 6. No matter which line in Figure 6 is, there is a higher $(\beta^*)^2$ value of core-shell composite particles than that of single phase solid particles only when ε_2 is fallen in the region of $\varepsilon_2 < 1$ or $\varepsilon_p < \varepsilon_{pi}$. If $\varepsilon_i > 100$, the interforce improvement is limited using a core material with $\varepsilon_2 < 1$ despite the single phase solid particle is changed to a core-shell structure (curves b and d). On the other hand, if the structure of the single phase particles with $\varepsilon_i < 100$ is changed to a core-shell structure, the value of b/a will apparently affect the improvement (curves a and c).

For the case of $\varepsilon_i > 1$, consider two extremes ε_2 , (i) $\varepsilon_2 \rightarrow 0$ (Route I_a), (ii) $\varepsilon_2 \rightarrow \infty$ (Route I_b):

(i) Route I_a: In the case of $\varepsilon_2 \rightarrow 0$, the core material is a metal. $M = -b^3/a^3$, obtained from Equation 10c, is substituted into Equation 10b to

produce Equation 11.

$$\alpha = \varepsilon_1 G_o \quad (11)$$

where

$$G_o = \frac{\left(1 + \frac{2a^3}{b^3}\right)}{\left(1 - \frac{a^3}{b^3}\right)} \quad (12)$$

When the shell thickness ($\delta=b-a$) is extremely thin, the correction term appearing in Equation 12 can be reduced to

$$G_o \approx \frac{b}{(b-a)} = \frac{b}{\delta} \quad (13)$$

Equation 11 is the same as that derived from Davis (1993). Since $b>a$ for the core-shell, the geometric correction term is greater than one, that is $G_o>1$. Therefore, if the structure of single phase solid particles with $\varepsilon_p/\varepsilon_f>1$ is changed to a core-shell structure and a higher dielectric material or a metallic material is used to replace the core (Figure 5), the ER fluid will have a larger α value and a higher $(\beta^*)^2$ than those of the single phase solid particles. The inter-particle forces are largely increased especially for the metal core. From Equation 13, we know that in the case of $\varepsilon_p/\varepsilon_f>1$, the thinner the shell, the larger the G_o is. This represents that the interforce becomes larger and the ER effect is much improved for particles with larger metal cores. Therefore, Davis claimed that metallic particle coated with an insulating layer could be used as an ideal ERF suspended particle. However, from our conclusions, the dielectric constant of the insulating layer must be higher than that of the surrounding medium. Otherwise, the inverse polarization will happen and results in lower yield stresses of an ERF. The experiments performed by Wen and Lu coated an oxide layer on aluminum particles to fabricate shell-core composite particles under high temperature [23]. The yield stress was not high either under low frequency field or under high frequency field and it reached only several Pascals.

In addition, the larger metal core will increase the particle mass. As a result, precipitation will appear and must be solved. Therefore, considering the conductivity and density, a hollow metal sphere with a thin shell is considered to solve this problem. In practice, it is difficult to make this structure during

processing. Furthermore, solving the precipitation problem should use the other scheme. Polymeric powders coated with a metal shell using a chemical method were made to replace the metal core. Because of the metal shell around the polymeric core, the electrical properties of the polymeric core are shielded. Thus, the bulk electrical properties of the multi-shell particle will not be affected. Therefore, considering the cost and the process, the polymeric material as a core is a suitable choice to adjust bulk density of particles.

(ii) Route I_b : In the case of $\varepsilon_2 \rightarrow \infty$, the single phase particle is nearly a hollow sphere. $M=2b^3/a^3$, obtained from Equation 10c, is substituted into Equation 10b producing Equation 14.

$$\alpha = \varepsilon_1 G_\infty \quad (14)$$

where

$$G_\infty = \frac{\left(2 - \frac{2a^3}{b^3}\right)}{\left(2 + \frac{a^3}{b^3}\right)} \quad (15)$$

The magnitude of the geometric correction term is greater than zero and smaller than one, $0<G_\infty<1$. If the structure of the single phase solid particles with $\varepsilon_p/\varepsilon_f>1$ is changed to a core-shell structure and the core material is replaced by a lower dielectric material or the core is kept empty forming a hollow sphere, the ER fluid will degrade and have a smaller α value and a lower inter-particle force $(\beta^*)^2$ than those of the single phase solid particles.

It should be emphasized that no matter what kind of material is chosen for the core, the value α^* can not be greater than one when $\varepsilon_f<1$. Otherwise, the inverse polarization will happen when the particles immerse in an ER suspension under the electric field and the yield stress of ERF can not be high enough. It is important that the core material improvement is strongly related to the right choice of the shell material.

Scheme II: Coating a shell on single phase solid particles

The second scheme is coating a shell with material B surrounding the single phase solid particle made of material A to form a core-shell particle as shown in route II of Figure 2.

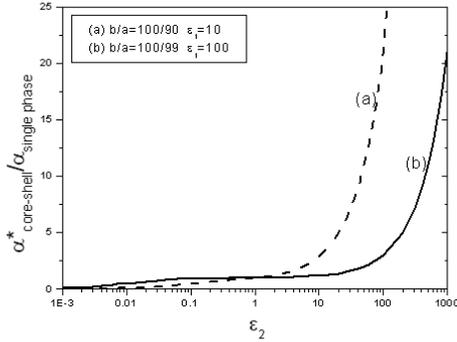


Fig. 7. $\frac{\alpha^*}{\alpha}$ as a function of ϵ_2 for a core-shell structural particle at different geometric conditions.

For the case of single phase solid particles, $\alpha = \epsilon_p / \epsilon_f$ expresses the dielectric mismatch while $\alpha^* = (\epsilon_p / \epsilon_f) \{ (M-2) / (M+1) \}$ expresses the dielectric mismatch for the core-shell particle case. The mismatch ratios of the core-shell particles to the single phase solid particles are $\frac{\alpha^*}{\alpha} = \epsilon_2 \left(\frac{M-2}{M+1} \right)$

which is different from the factor $\left(\frac{M-2}{M+1} \right)$ appeared in Route I. The relations of α^*/α versus ϵ_2 for particles with a core-shell structure and for particles with a single phase structure were computed. The results are shown in Figure 7 for different dielectric ratios, $\epsilon_f = 10$ and 100 , under the conditions of $b/a = 100/90$ and $100/99$. The ER improvement is effective only if $\epsilon_2 > 1$. The larger the ϵ_2 , the better the improvement is. The thicker the coated shell, the better the improvement is. In brief, as long as the dielectric constant of the shell material is higher than that of the core material, the ER improvement will be effective as a shell material is coated on a single phase solid particle and the core material is kept the same as the single phase solid particle. Furthermore, the dielectric constant of the shell portion must be higher than that of the host liquid.

IV. CONCLUSIONS

A set of theoretical equations has been derived that satisfies Laplace's equation for

particles with a core-shell structure dispersed in a host liquid. These equations were used to predict the inter-particle force based on the dielectric polarized model. The computational results revealed that the larger the interforces the larger the particle/liquid dielectric mismatch ratio (β). In order to avoid the inverse polarization, the dielectric ratio of the solid particle to the host liquid should be greater than one for the single phase solid particles. While for the core-shell particles, the dielectric ratio of the shell material to the host liquid should be greater than one.

The interface between particles is improved, if the single solid phase is replaced by a core-shell structure. Under the conditions of $\epsilon_p / \epsilon_f > 1$, no matter changing the core material of core-shell particles or coating a shell on single phase solid particles, the dielectric constant of the replacing material or coating material must be higher than that of the material of the original single phase solid particles. Especially the metal core is the best choice. However, the larger metal core will increase the particle weight. As a result, avoiding the precipitation problem, coating a metal shell on the polymeric powders will be a better solution. The polymer core is used to replace the metal core bringing about the weight reduction of particles. The thickness of outermost shell of the multi-shell particle is considered to be as thin as possible in order to have better ER characteristics. Finally, this paper adopted a simple model not only explaining experimental results but also as a reference of fabrication of suspended particles in an electrorheological fluid.

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