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<th>Title</th>
<th>Exponential formula for computing effective viscosity</th>
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<td>Author(s)</td>
<td>Cheng, Nian-Sheng; Law, Adrian Wing-Keung</td>
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<tr>
<td>Citation</td>
<td>Cheng, N. S. &amp; Law, A. W. K. (2003). Exponential</td>
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<td>formula for computing effective viscosity. Powder</td>
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<td>Technology, 129(1-3), 156–160.</td>
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<td>The published version is available at: [DOI:</td>
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<td><a href="http://dx.doi.org/10.1016/S0032-5910(02)00274-7">http://dx.doi.org/10.1016/S0032-5910(02)00274-7</a>].</td>
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EXponential FOrmula For COmputing EffeCTive ViSCOsiTY

Nian-Sheng Cheng¹ and Adrian Wing-Keung Law²

ABSTRACT: An exponential model is proposed in this paper for evaluating the effective viscosity of a particle-fluid mixture. First, the theoretical consideration is restricted to the dilute condition without the effects of dynamic particle interactions and fluid turbulence. This leads to a power series expressed in terms of particle concentration, which can be viewed as an extension of the Einstein’s formula. The derived expression is then modified by including indirectly effects of inter-particle collisions, fluid turbulence and random motion of the particles, which can cause the effective viscosity to be increased significantly with increasing particle fraction in the mixture. Finally comparisons are made between the present study and various theoretical and empirical results available in the literature and satisfactory agreement is observed.

Keywords: effective viscosity, concentration of particles, suspension, two-phase flow, particle interaction, dilute suspension

INTRODUCTION

The phenomenon of solid particles suspended in fluid is of key interest in hydraulic engineering such as sediment transport in rivers and slurry transport in pipelines. Similar two-phase suspensions can also be found in industrial applications such

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as the processing of cement, composite materials and foodstuffs. As an important macroscopic property of these suspensions, the effective viscosity is usually required in the analysis of the related transport processes.

It is well known that the earliest theoretical work on the effective viscosity was due to Einstein (1906) whose derivation led to the effective viscosity to be linearly related to the particle concentration as follows:

\[ \mu_r = 1 + 2.5\phi_p \]  

(1)

where \( \mu_r = \mu_m/\mu_f \) = relative viscosity defined as the ratio of the effective viscosity of the particle-fluid mixture, \( \mu_m \), to the viscosity of fluid, \( \mu_f \), and \( \phi_p \) = volumetric concentration of the particles. This expression is exact when the viscous effect is dominant so that the creeping flow equations can be applied at the particle level. Since the influence of particle interaction is not considered, (1) is only applicable to suspensions with low particle concentrations, say, less than 2% (Acrivos 1995).

Following Einstein’s work, numerous expressions have been proposed to extend the range of validity to higher concentrations. They are either theoretical expansions of (1) to higher order in \( \phi_p \), or empirical expressions that were obtained based on experimental data. The theoretical expansions are usually expressed in the form of a power series,

\[ \mu_r = 1 + k_1 \phi_p + k_2 \phi_p^2 + k_3 \phi_p^3 + \cdots \]  

(2)

where \( k_1, k_2, k_3 = \) coefficients. Evaluation of these coefficients other than \( k_1 \) requires the formulation of particle interactions, which is rather difficult. This is one of the reasons that only the theoretical values of the coefficients related to lower order in \( \phi_p \), like \( k_2 \) and
k₃, can be found in the literature so far. Even for these lower order coefficients, the calculations are available only for idealized cases when the particle arrangement or its statistic properties are simple. An example for determining the k₂-value was presented by Batchelor and Green (1972), who reported that the coefficient k₂ was equal to 7.6 for a suspension undergoing a pure straining motion. It should be noted however that Batchelor and Green introduced a rough interpolation in their derivation, which led to an inaccuracy in the k₂-value obtained. A re-calculation under the same condition was conducted by Kim and Karrila (1991), leading to the k₂-value to be changed to 6.95. Other k₂-values have been reviewed by Happel and Brenner (1986). Furthermore, Thomas and Muthukumar (1991b) found the third order coefficient, k₃, to be 6.40 by applying the multiple scattering theory to the evaluation of the hydrodynamic interaction of three spheres.

Following these theoretical attempts, it can be expected that more complicated configurations of the particle arrangements will lead to further difficulties in the mathematical formulation. On the other hand, particles suspended in fluid should in fact be distributed in a random manner so that their arrangement cannot be fitted to a simple configuration or described by a simple distribution function. Therefore, although an idealized particle arrangement may enable a theoretical evaluation of the k-coefficients in higher order, the applicability of the effective viscosity so derived is limited in practice. Comparisons with experimental results show that the existing theoretical expressions are only applicable to those conditions with low concentration up to 10% (Barnes et al. 1989).
In addition to the theoretical expressions, various empirical relationships have been proposed to evaluate the effective viscosity of a suspension with higher concentration. An early survey of such relationships made by Rutgers (1962) showed that large discrepancies exist among the different relationships. Acrivos (1995) claimed that the discrepancies could partially be attributed to the viscometric measurements that were normally subjected to shear-induced particle diffusion.

Table 1 shows some typical examples of the existing empirical formulas, where $\phi_{p\text{max}}$ = maximum particle concentration and $\mu_{\text{in}}$ = intrinsic viscosity. It is noted that for those relationships where the so-called maximum concentration is included as a parameter, the effective viscosity approaches infinity when the concentration is equal to the maximum value. This may not be physically reasonable. Strictly speaking, there are only two extreme conditions that are meaningful for the effective viscosity. The first condition is a suspension without particles, implying that the effective viscosity is the same as the fluid viscosity. The other is a suspension without fluid, which would then theoretically behave as a solid with infinite viscosity. On the other hand, only for particles with regular shapes can the maximum concentration be mathematically determined for a given packing arrangement. For example, the maximum concentration can reach 1.0 for cubes packed face-to-face and 0.74 for spheres with the closest hexagonal packing arrangement. With irregular particles, the maximum value varies markedly even though the particle size is uniform. As a matter of fact, the maximum concentration and intrinsic viscosity were often used as two empirical parameters in fitting experimental data to an empirical formula (e.g., Leighton and Acrivos 1987, Barnes et al. 1989).
Other than the traditional approaches mentioned above, Fan and Boccaccini (1996) studied the effective viscosity using topological transformation. Their results showed that the effective viscosity increases with increasing continuity of the solid particles. This suggests the existence of two limiting cases. One is the minimum effective viscosity when the particles are completely discontinuous, and the other is the maximum effective viscosity when the particles are completely continuous. Their results should be viewed as qualitative since a number of topological parameters were defined for characterizing the microstructure of the two-phase system, of which some are not experimentally deterministic or even not physically clear.

This study starts with a derivation of the effective viscosity for the dilute condition without dynamic particle interaction. The expression of the effective viscosity so obtained recovers the Einstein's formula. The expression is further modified to be applicable for high particle concentrations. Comparisons of the present study with other empirical relationships are finally provided.

**DERIVATION**

**Effective Viscosity for Dilute Conditions**

The following approach illustrates an iterative concept, based on the Einstein’s formula, that is simple in implementation yet retains the basic physical representation of the situation. The objective is to derive the effective viscosity in the dilute condition but
with the particle concentration beyond the range of validity of the Einstein formula. Therefore, the inter-particle collision and effects of the fluid turbulence and particle random motion are assumed to be insignificant.

Consider a suspension made of clean fluid with uniformly distributed particles, whose volumes are \( v_f \) and \( v_p \), respectively. First, divide \( v_p \) into \( n \) sub-volumes, i.e.

\[
v_p = \Delta v_{p,1} + \Delta v_{p,2} + \Delta v_{p,3} + \cdots + \Delta v_{p,n}
\]  

(3)

With this division, a series of slightly different suspension samples can be constructed with progressively increasing particle concentration, as sketched in Fig. 1. It is noted that although the particle volume varies in the different samples, the fluid volume remains constant. For simplicity, all the particle volume increments can be set to be equal, i.e., \( \Delta v_{p,i} = \Delta v_p \) for \( i = 1, 2, \ldots, n \). Hence, the sample volumes are \( v_f + \Delta v_p, v_f + 2\Delta v_p, \ldots, v_f + n\Delta v_p \), respectively.

The effective viscosities of the samples are denoted as \( \mu_{m,1}, \mu_{m,2}, \ldots, \mu_{m,n} \), respectively. Their relationship can be conceptualized as follows. Consider the \((i-1)\)th sample with the effective viscosity, \( \mu_{m,i-1} \). If an additional volume of particles of \( \Delta v_p \) is further dispersed in this sample, it will then be changed to the \( i \)th sample with the effective viscosity increased to \( \mu_{m,i} \). Since \( \Delta v_p \) is arbitrary, it can be chosen in such a manner that \( \Delta v_p / (v_f + i\Delta v_p) \). for \( i = 1, 2, \ldots, n \), is small within the range of validity of the Einstein’s equation. With these considerations, it is expected that (1) is applicable to the \( i \)th suspension in the form

\[
\mu_{m,i} = \mu_{m,i-1} \left(1 + 2.5\phi_{pr,i}\right)
\]

(4)

where \( \phi_{pr,i} = i^{th} \) relative volumetric concentration defined as
Substituting (5) into (4) yields

\[ \mu_{m,i} = \mu_{m,i-1} \left( 1 + 2.5 \frac{\Delta v_p}{v_f + i\Delta v_p} \right) \]  

(6)

Note that \( \mu_{m,0} = \mu_f \). Applying (6) repeatedly from \( i = 1 \) to \( i = n \), we can get

\[ \mu_{m,n} = \mu_f \prod_{i=1}^{n} \left( 1 + \frac{2.5 \Delta v_p}{v_f + i\Delta v_p} \right) \]  

(7)

Furthermore, since \( \Delta v_p = v_p/n \) and \( v_p/(v_p + v_f) = \phi_p \), (7) can be changed to

\[ \mu_{m,n} = \mu_f \prod_{i=1}^{n} \left( 1 + \frac{2.5\phi_p}{n(1-\phi_p) + i\phi_p} \right) \]  

(8)

If \( n \) is large, the effective viscosity of the suspension with the volumetric particle concentration \( \phi_p \) can be expressed as

\[ \mu_m = \mu_f \lim_{n \to \infty} \left[ \prod_{i=1}^{n} \left( 1 + \frac{2.5\phi_p}{n(1-\phi_p) + i\phi_p} \right) \right] \]  

(9)

Using the Gamma function and its recursive property, (9) can be converted to

\[ \mu_e = \lim_{n \to \infty} \left[ \frac{\Gamma \left( \frac{n}{\phi_p} + 3.5 \right) \Gamma \left( \frac{1-\phi_p}{\phi_p} + 1 \right)}{\Gamma \left( \frac{n}{\phi_p} + 1 \right) \Gamma \left( \frac{1-\phi_p}{\phi_p} + 3.5 \right)} \right] \]  

(10)

Furthermore, using the following relationship (Abramowitz and Stegun 1970)

\[ \lim_{x \to \infty} \left[ \frac{\Gamma(x+a)}{\Gamma(x+b)} \right] = \lim_{x \to \infty} x^{a-b} \]  

(11)

where \( a, b = \) parameters, (10) can finally be simplified to
\[ \mu_r = \lim_{n \to \infty} \left( \frac{n^{1.5}}{\Phi_p} \right)^{2.5} \left( n \left( 1 - \frac{\Phi_p}{\Phi} \right) \right)^{-2.5} = (1 - \Phi_p)^{-2.5} = \Phi_f^{-2.5} \]  

(12)

where \( \phi_f = 1 - \phi_p \) = volumetric fraction of fluid. The above derivation is based the iterative concept without any empiricism, hence the resulting formula (12) should be considered as analytically vigorous and a direct extension of (1). The formula can further be expanded in a power series as

\[ \mu_r = 1 + \frac{5}{2} \Phi_p + \frac{35}{8} \Phi_p^2 + \frac{105}{16} \Phi_p^3 + \frac{1155}{128} \Phi_p^4 + \frac{3003}{256} \Phi_p^5 + \cdots \]  

(13)

This gives \( k_2 = 4.38 \) and \( k_3 = 6.56 \), which are very close to the coefficients computed by Thomas and Muthukumar (1991a, 1991b), as discussed previously.

As the dynamic effects of particles and fluid are not included, (12) can only be applied to dilute suspensions. If the particle concentration is increased, inter-particle collisions and random motion of particles are unavoidable, and thus the effective viscosity is also increased. This phenomenon can be illustrated effectively by introducing different stress components for fluid and solid phases, respectively, as detailed in Appendix I. Theoretically, the effective viscosity subjected to the dynamic effects in the non-dilute conditions can be obtained if the different stresses included in (24) can be formulated. Unfortunately, this is almost impossible as the information related to the particle collision and kinetic stress is still very limited in the literature. Alternatively, an exponential model is proposed in the following to extend (12) for the case of higher particle concentrations.
**Exponential Model**

There are a large number of measurements available in the literature for the effective viscosity. It is known that a considerable discrepancy exists in the measured viscosities at higher particle fractions reported by different investigators, although often each of their own data set showed little scatter (Acrivos 1995, Fan and Boccaccini 1996). The discrepancy may depend on the property of particles, shear rate and mechanism of the viscometer.

For a specific suspension, however, the relative viscosity generally varies with increasing fluid fraction described as follows. First, the relative viscosity declines rapidly with increasing fluid fraction, $\phi_f$, for small $\phi_f$-values. This is then followed by a gentle reduction if $\phi_f$ is further increased. For the limiting condition of $\phi_f = 1$, the relative viscosity would then be equal to one. Such variations can be illustrated in Fig. 2, for example, by an empirical relationship which was obtained for a suspension comprised of 46 µm polystyrene spheres (Leighton and Acrivos 1987). Also superimposed in the figure is (12), which serves as an asymptote to the measurements for very high $\phi_f$-values but deviates from the empirical relationship, as expected, if $\phi_f$ is reduced.

Fig. 2 shows that the slope of the trendline fitted to the experimental data, when plotted on the logarithmic scale, always decreases with increasing fluid fraction, approaching 2.5 for the dilute condition. Mathematically, as a first approximation, this can be expressed as

$$\frac{d \ln \mu_r}{d \ln \phi_f} = -\frac{2.5}{\phi_f^5}$$  \hspace{1cm} (14)
where $\beta = \text{exponent}$. Integration of (14) with respect to $\phi_f$ leads to

$$\mu_r = \alpha \exp \left( \frac{2.5}{\beta \phi_f^\beta} \right)$$

(15)

Since $\mu_r = 1$ for $\phi_f = 1$,

$$\alpha = \exp \left( - \frac{2.5}{\beta} \right)$$

(16)

Substituting (16) into (15) yields

$$\mu_r = \exp \left[ \frac{2.5}{\beta} \left( \frac{1}{\phi_f^\beta} - 1 \right) \right]$$

(17)

or

$$\mu_r = \exp \left[ \frac{2.5}{\beta} \left( \frac{1}{(1-\phi_p)\beta} - 1 \right) \right]$$

(18)

Fig. 2 shows a family of curves plotted according to (17). It is noted that with increasing $\beta$-values, the $\mu_r - \phi_f$ relationship deviates gradually from $\mu_r = \phi_f^{-2.5}$, and for the same fluid fraction, $\mu_r$ increases with increasing $\beta$-values.

**COMPARISON WITH PREVIOUS STUDIES**

With different $\beta$-values, (18) can be used to represent various empirical relationships included in Table 1, which were derived largely based on experimental data. For example, the formula given by Mooney (1951) for $\phi_{p_{\text{max}}} = 0.74$ and that reported by Leighton and Acrivos (1987) can be well approximated by (18) with $\beta = 2$; while Barnes’
expression for the high shear condition is close to (18) for $\beta = 0.95$. More comparisons are given in Fig. 3, where the relative viscosity is plotted against the concentration of particles, and the corresponding $\beta$-values are found varying from 0.95 to 3.9.

Clearly, for $\beta = 0$, (19) reduces to (13). Comparing (19) with (2) yields that all the $k$-coefficients except for $k_1 = 2.5$ are functions of $\beta$. The computed results included in Table 2 show that $k_2 = 5.63 \sim 9.38$, $k_3 = 11.35 \sim 30.73$, $k_4 = 21.32 \sim 93.82$ and $k_5 = 37.95 \sim 272.79$ if $\beta$ varies from 1 to 4.

Also included in Table 2 are various values of the coefficients from $k_1$ to $k_5$, which were obtained theoretically or empirically by previous researchers. The coefficients for the formula reported in Thomas (1965) were calculated by expanding his formula into a series, while those for Ward’s formula (see Graf 1977) by setting $k_1 = 2.5$. Table 2 shows that there are marked differences among the various values for each coefficient, including those obtained in the present study. However, it is interesting to note that the computed coefficients of $k_2$ to $k_5$ for $\beta = 2$ are very close to those reported by Ward (1955, see Graf 1977) who suggested the following expression

$$\mu = 1 + k\phi_p + (k\phi_p)^2 + (k\phi_p)^3 + (k\phi_p)^4 + \ldots$$

(20)
to be fitted to the experimental data for the concentration up to 35%, where \( k = 2.5 \) for spheres.

CONCLUSIONS

The objective of this study was not to develop another empirical formula to represent the existing experimental data on top of other dozens of empirical relationships available in the literature. Rather, we demonstrate an approach to calculate the effective viscosity for a particle suspension that is simple to implement yet maintains the underlying physical representation of the situation. The results obtained indicate that it is not necessary to involve the so-called maximum particle concentration, which is closely related to the randomness of particle parking, in computing the effective viscosity. For the condition without dynamic particle interactions and turbulence effects, the derived formula serves as an extension of the well-known Einstein’s expression. This formula, after further modification, is able to match well with the empirical relationships reported previously. The analytical results obtained can also be given in the form of power series for comparison purposes.
APPENDIX I. STRESS ANALYSIS

The bulk stress in a two-phase suspension is composed of two components, one due to the fluid phase and the other due to the particle phase. Simply, it can be expressed as

\[ \tau_m = \phi_f \tau_f + \phi_p \tau_p \] (21)

where \( \tau_f \) = fluid phase stress and \( \tau_p \) = particle phase stress. Furthermore, it is known that the fluid phase stress can be decomposed as

\[ \tau_f = \tau_f^v + \tau_f^l \] (22)

where \( \tau_f^v \) = fluid viscous stress and \( \tau_f^l \) = fluid turbulence stress. In comparison, the particle phase stress can be divided into a static component, which is only associated with the stationary existence of the particles in the fluid, and a dynamic component, which is related to the relative motion among the particles or particle interaction. The relative motion causes velocity fluctuations of the particles as well as inter-particle collisions. For convenience, the particle phase stress can also be divided into three components as follows (Hwang and Shen 1989)

\[ \tau_p = \tau_p^c + \tau_p^k + \tau_p^p \] (23)

where \( \tau_p^c \) = particle collision stress, \( \tau_p^k \) = particle kinetic stress and \( \tau_p^p \) = particle presence stress. The collision stress is the rate of momentum transfer due to the collisions between the particles. The kinetic stress is the rate of momentum transfer caused by the random motion of the particles, and thus can also be called the particle turbulence stress. The particle presence stress is not due to the relative motion among the particles. Rather, it is
a result of the existence of the particles in the fluid. The volume fraction of the particles alters the local flow pattern of the fluid and thus affects the bulk rheological property.

Substituting (22) and (23) into (21), one gets

\[ \tau_m = \phi_f (\tau_f + \tau_f^l) + \phi_p (\tau_p^c + \tau_p^k + \tau_p^p) \]  

(24)

Eq. (24) shows that for a suspension with a specified concentration of particles, the stress minimizes if the contributions by the fluid turbulence, inter-particle collision and particle random motion can be neglected, i.e., \( \tau_f^l, \tau_p^c \) and \( \tau_p^k \) are approximately equal to zero. This condition in effect is implied in deriving (1) for the case of dilute suspensions (Hwang and Shen 1989), where the particle presence stress can simply be obtained by integrating the Stokes stress. With an increase in the particle fraction, particle interactions become significant and at least \( \tau_p^c \) and \( \tau_p^k \) cannot be ignored, thus the stress increases. For the same strain rate, the higher the stress, the larger the effective viscosity of the suspension.

Theoretically, the effective viscosity can be derived by considering the various stress components included in (24). However, since general information related to the collision and kinetic stresses is still not available, difficulties arise in order to further formulate the different stress components.
APPENDIX II. REFERENCES


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APPENDIX III. NOTATION

The following symbols are used in this paper:

- \( a \) = parameter;
- \( b \) = parameter;
- \( k \) = coefficient;
- \( k_i \) = coefficient (\( i = 1, 2, 3, \ldots \));
- \( n \) = total number of suspension samples;
- \( \tau_f \) = fluid phase stress;
- \( \tau_{f}^{i} \) = fluid turbulence stress;
- \( \tau_{f}^{v} \) = fluid viscous stress;
- \( \tau_m \) = bulk stress tensor;
- \( \tau_p \) = particle phase stress;
- \( \tau_{p}^{c} \) = particle collision stress;
- \( \tau_{p}^{k} \) = particle kinetic stress;
- \( \tau_{p}^{p} \) = particle presence stress;
- \( v_f \) = volume of fluid;
- \( v_p \) = volume of particles;
- \( \alpha \) = coefficient;
- \( \beta \) = exponent;
- \( \mu_f \) = viscosity of fluid;
\( \mu_n \) = intrinsic viscosity;
\( \mu_m \) = effective viscosity of particle-fluid mixture;
\( \mu_{m,i} \) = effective viscosity of the ith suspension sample (i = 1, 2, ..., n);
\( \mu_r \) = relative viscosity = \( \mu_n / \mu_r \);
\( \phi_f \) = volumetric fraction of fluid;
\( \phi_p \) = volumetric concentration of particles;
\( \phi_{p\text{max}} \) = maximum volumetric concentration;
\( \phi_{pr,i} \) = ith relative volumetric concentration;
\( \Delta v_{p,i} \) = increment of volume of particles (i = 1, 2, ..., n).
Table 1. Empirical Formulas for Effective Viscosity

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<tr>
<th>Researchers</th>
<th>( \frac{\mu_m}{\mu} )</th>
<th>( \mu_n )</th>
<th>( \phi_{p_{\text{max}}} )</th>
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<td>Mooney (1951)</td>
<td>[ \exp\left( \frac{2.5\phi_p}{1 - \phi_p/\phi_{p_{\text{max}}}} \right) ]</td>
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<td>0.52~</td>
</tr>
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<td>Thomas (1965)</td>
<td>( 1 + 2.5\phi_p + 10.05\phi_p^2 + 0.00273 \exp(16.6\phi_p) )</td>
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<td>Metzner (1985)</td>
<td>[ \left( 1 - \frac{\phi_p}{\phi_{p_{\text{max}}}} \right)^{-2} ]</td>
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<tr>
<td>Leighton and Acrivos (1987)</td>
<td>[ \left( 1 + \frac{0.5\mu_n\phi_p}{1 - \phi_p/\phi_{p_{\text{max}}}} \right)^2 ]</td>
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<td>0.58</td>
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<td>Barnes et al. (1989)</td>
<td>[ \left( 1 - \frac{\phi_p}{\phi_{p_{\text{max}}}} \right)^{-\mu_m\phi_{p_{\text{max}}}} ]</td>
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Table 2. Values of Coefficients included in Eq. (2)

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<td>5.63</td>
<td>11.35</td>
<td>21.32</td>
<td>37.95</td>
</tr>
<tr>
<td>(\beta = 2)</td>
<td>2.5</td>
<td>6.88</td>
<td>16.98</td>
<td>39.13</td>
<td>85.66</td>
</tr>
<tr>
<td>(\beta = 3)</td>
<td>2.5</td>
<td>8.13</td>
<td>23.44</td>
<td>63.09</td>
<td>161.54</td>
</tr>
<tr>
<td>(\beta = 4)</td>
<td>2.5</td>
<td>9.38</td>
<td>30.73</td>
<td>93.82</td>
<td>272.79</td>
</tr>
</tbody>
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Captions for Figures

Fig. 1. Sketch of Suspension Series

Fig. 2. Effective Viscosity Decreases with Increasing Fluid Fractions

Fig. 3. Variations of Effective Viscosity with $\beta$ as a Parameter

Fig. 4. Comparisons with Previous Empirical Relationships
\[ \mu_{m,i} = \mu_{m,1} \]

\[ \mu_{m,2} \]

\[ \mu_{m,i-1} \]

\[ \mu_{m,i} \]

\[ \mu_{m,n} \]

\[ \phi_{pr,i} = \frac{\Delta v_p}{v_f + \Delta v_p} \]

\[ \frac{\Delta v_p}{v_f + 2\Delta v_p} \]

\[ \frac{\Delta v_p}{v_f + (i-1)\Delta v_p} \]

\[ \frac{\Delta v_p}{v_f + i\Delta v_p} \]

\[ \frac{\Delta v_p}{v_f + n\Delta v_p} \]

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