

Effective viscosity of bidisperse suspensions

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(Received December 31, 2004; final revision received March 5, 2005)

Abstract

We determine the effective viscosity of suspensions with bidisperse particle size distribution by modifying an effective-medium theory that was proposed by Acrivos and Chang (1987) for monodisperse suspensions. The modified theory uses a simple model that captures some important effects of multi-particle hydrodynamic interactions. The modifications are described in detail in the present study. Estimations of effective viscosity by the modified theory are compared with the results of prior work for monodisperse and bidisperse suspensions. It is shown that the estimations agree very well with experimental or other calculated results up to approximately 0.45 of normalized particle volume fraction which is the ratio of volume fraction to the maximum volume fraction of particles for bidisperse suspensions.

Keywords : effective-medium theory, effective viscosity, bidisperse suspensions, multi-particle hydrodynamic interaction, conditional ensemble average

1. Introduction

Suspensions have been a long-standing subject in many theoretical and experimental studies for understanding physics of natural and industrial systems consisting of fluids and particles in various physical forms. In particular, there have been a lot of efforts to understand the microstructures and hence to determine the macroscopic rheological properties of suspensions. Prediction of suspension viscosity based on the calculation of hydrodynamic interactions among particles has been an important issue in the area of fluid mechanics. An interesting case can be suspensions with bidisperse or polydisperse size distribution of particles. In present study we estimate effective viscosity of bidisperse suspensions using a simple model and compare the estimations with experimental or other theoretical results.

Due to the fundamental and practical importance, a number of investigations have been performed to understand rheological behavior of bidisperse suspensions and hence to determine the effective viscosity of the suspensions. Chong *et al.* (1971) have extensively investigated the rheological behavior of multimodal suspensions of glass spheres. Poslinski *et al.* (1988) have experimentally determined the effective viscosity of bidisperse suspensions for a wide range of volume fractions of particles. The effective

viscosity of monodisperse and bidisperse suspensions for very high volume fractions of particles has been obtained by Shapiro and Probstein (1992) to determine the random close-packing fractions of the suspensions. Chang and Powell (1993) have simulated dynamics of monolayer of suspensions with bidisperse distribution of hard spheres using a numerical technique that was originally developed by Brady and Bossis (1988) with the basis on Stoksonian dynamics. Numerical simulation methods for computing hydrodynamic interactions among hard spheres have been also developed by Ladd (1990) and Mo and Sangani (1994) for monodisperse suspensions and by Koo and Sangani (2002) for bidisperse suspensions. These complicated numerical simulations have been often compared with effective-medium theories based on simple models. An effective-medium theory has been proposed by Acrivos and Chang in a series of their papers (Chang and Acrivos, 1986a; 1986b; Acrivos and Chang, 1987). This theory based on pair probability, i.e. probability of finding a particle at a position given a particle at origin, models random monodisperse suspensions as a particle at origin surrounded by an effective-medium in which its properties such as density and viscosity change continuously depending on pair probability. This theory has been shown to be in a good agreement with experimental results for a problem of heat conduction from a heated sphere placed in a packed bed with monodisperse passive spheres (Acrivos and Chang, 1987).

We describe modifications of this theory for estimating

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the effective properties of bidisperse suspensions in detail in the present study. The modifications are aimed at providing a simple method to predict effective viscosity of bidisperse suspensions. The estimations by the modified theory are compared with the experimental or calculated results.

2. Theory

2.1. Effective-medium theory

Effective-medium theories estimate conditionally averaged fields and hence the properties of suspensions by solving suitably averaged equations for a relatively simple model that captures some of the important multi-particle effects. The first step in developing the theory is to derive an equation for the conditionally averaged velocity and to introduce appropriate closures, and the second step is the construction of model to evaluate the unknown constants appearing in the closures.

We consider a suspension of rigid spherical particles of radius a in a Newtonian fluid. When the Reynolds number based on the particle radius is small, the fluid motion satisfies

$$\frac{\partial \sigma_{ij}}{\partial x_j} + \rho_f g_i = 0 \quad (1)$$

where σ_{ij} is stress at point \mathbf{x} in the fluid, ρ_f the density of the fluid, and g_i the gravitational acceleration. The stress inside the particle satisfies a similar equation with ρ_f replaced by the particle density ρ_p . Ensemble-averaging these equations subject to the presence of a particle with its center at origin, $\mathbf{0}$, yields

$$\frac{\partial \langle \sigma_{ij} \rangle_c(\mathbf{x}|\mathbf{0})}{\partial x_j} + \rho(\mathbf{x}) g_i = 0 \quad (2)$$

with

$$\rho(\mathbf{x}) = \rho_f + (\rho_p - \rho_f) \langle \chi \rangle_c(\mathbf{x}|\mathbf{0}). \quad (3)$$

Here, χ is a particle phase indicator function whose value is unity when \mathbf{x} lies inside a particle and zero otherwise. $\langle \chi \rangle_c(\mathbf{x}|\mathbf{0})$ is conditional average of χ that can be written as

$$\langle \chi \rangle_c(\mathbf{x}|\mathbf{0}) = \int_{|\mathbf{x}-\mathbf{x}'| \leq a} P(\mathbf{x}'|\mathbf{0}) dV_{\mathbf{x}'} \quad (4)$$

where $P(\mathbf{x}'|\mathbf{0})$ is probability density for finding a particle with its center at \mathbf{x}' given the presence of a particle at the origin. Note that χ approaches ϕ as $|\mathbf{x}| \rightarrow \infty$. For suspensions with isotropic pair probability density a closure relation for the stress is introduced

$$\langle \sigma_{ij} \rangle_c = -\langle p \rangle_c \delta_{ij} + \eta(\mathbf{x}) \left[\frac{\partial \langle u_i \rangle_c}{\partial x_j} + \frac{\partial \langle u_j \rangle_c}{\partial x_i} \right] \quad (5)$$

where $\langle p \rangle_c$ is the conditionally averaged pressure and $\eta(\mathbf{x})$ is viscosity of the suspension at \mathbf{x} . The conditionally aver-

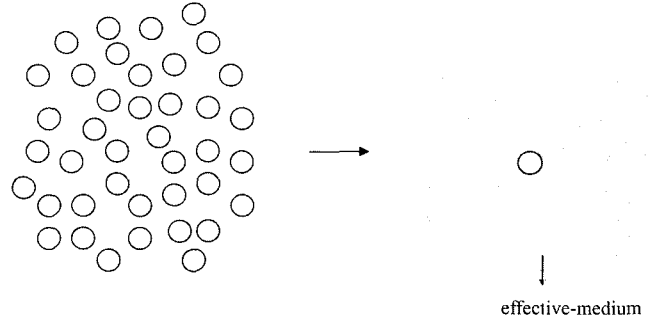


Fig. 1. Schematic diagram of effective-medium model.

aged pressure and velocity are required to approach the unconditional averages as $|\mathbf{x}| \rightarrow \infty$, respectively.

In order to solve the conditionally averaged equations above, we use a simple effective-medium model proposed by Acrivos and Chang (1987). This model is represented by a test particle surrounded by an effective-medium (Fig. 1) in which the properties such as density and viscosity are allowed to vary continuously. For example, the density is taken as Eq. (3). Likewise the effective viscosity of the medium is taken as

$$\eta(\mathbf{x}) = \eta_f + (\eta^* - \eta_f) \langle \chi \rangle_c(\mathbf{x}|\mathbf{0}) / \phi \quad (6)$$

where η_f and η^* are the viscosities of fluid and effective-medium, respectively. For suspensions in which the pair probability density is independent of the orientation of the pair, volume integral in Eq. (4) can be reduced using geometrical consideration shown in Fig. 2 to integration over R ;

$$\langle \chi \rangle_c(\mathbf{x}|\mathbf{0}) = n \pi \int_{r-a}^{r+a} g(R) (2R - R^2/r - r + a^2/r) R dR \quad (7)$$

where g is radial distribution function for monodisperse hard spheres.

Extending this model to bidisperse suspensions, Eq. (1) subject to the presence of a particle of radius a_1 with its center at origin is given by

$$\frac{\partial \langle \sigma_{ij} \rangle_c(\mathbf{x}|\mathbf{0}, a_1)}{\partial x_j} + \left[\rho_f + \sum_{k=1}^2 (\rho_{p,k} - \rho_f) \langle \chi_k \rangle_c(\mathbf{x}|\mathbf{0}, a_1) \right] g_i = 0 \quad (8)$$

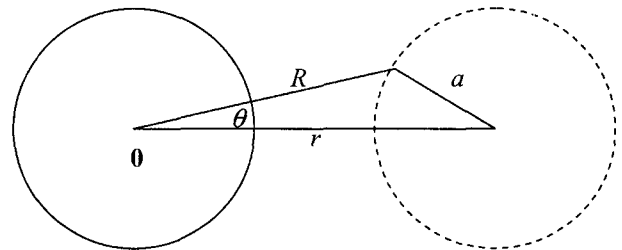


Fig. 2. Geometry for the integration in Eq. (4).

where χ_k ($k = 1, 2$) are the indicator functions for k -species particles. In viscosity problem, $\rho_p = \rho_f$. Thus $\rho_f g_i$ remains as the second term in Eq. (8).

Likewise Eq. (7) is rewritten as

$$\langle \chi_i \rangle_c(x|0, a_j) = n_i \pi \int_{r-a_j}^{r+a_j} g_{ij}(R) (2R - R^2/r - r + a_j^2/r) R dR \quad (9)$$

where g_{ij} is radial distribution function for a species i given a species j at origin.

The radial distribution function for binary mixtures of hard spheres g_{ij} is obtained by solving the generalized Percus-Yevick equation. Lebowitz (1964) has given the expressions for g_{ij} in terms of Laplace transforms. These need to be inverted to determine g_{ij} as functions of r for our calculation.

Throop and Bearman (1965) have used a numerical method for inverting the Laplace transforms. Later, Leonard *et al.* (1971) provided an explicit expression for g_{ij} using the inversion procedure described by Throop and Bearman (1965). However, the formulas given by these later investigators are incorrect.

We, therefore, followed the calculation procedure for g_{ij} given by Throop and Bearman (1965) and obtained the expressions for g_{ij} in explicit forms. The formulas are given in Appendix. Alternatively, a Monte Carlo simulation can be also applied to obtain the radial distribution function for bidisperse hard spheres as given by Chun *et al.* (2004).

2.2. Effective-medium calculations

As mentioned earlier, the conditionally averaged velocity and hence effective viscosity were determined by solving the effective-medium equation. The conditionally averaged velocity satisfies

$$\nabla \cdot \{ \eta(r) [\nabla \mathbf{u} + (\nabla \mathbf{u})^+] \} + \rho(r) \mathbf{g} - \nabla p = 0 \quad (10)$$

where \mathbf{u} and p are the conditionally averaged velocity and pressure respectively.

And $\rho(r)$, density of effective-medium, is also a function of the distance r from origin as is the viscosity. In this calculation the mean flow is chosen such that the conditionally averaged velocity is axisymmetric around x_1 axis. Thus it is possible to introduce a stream function to simplify the equations of motion. The stream function can be expressed as a function of r times a function of μ

$$\psi = f_n(r) Q_n(\mu) \quad (11)$$

where $\mu = \cos \theta$, θ being the polar angle measured from the x_1 axis, and Q_n is the integral of the Legendre function (see, e.g., Leal, 1992) For viscosity calculation, n equals 2 but this subscript is omitted for convenience. The function f must be determined by numerical integration of the equations of motion with the boundary conditions of no slip at the surface of the particle at origin and mean flow at infinity from the origin. The equation of motion in this problem

is written as

$$f^{(4)} + \frac{2\eta'}{\eta} f''' + \left(\frac{\eta''}{\eta} - \frac{2\eta'}{r\eta} - \frac{12}{r^2} \right) f'' - \left(\frac{2\eta''}{r\eta} + \frac{10\eta'}{r^2\eta} - \frac{24}{r^3} \right) f' + \left(\frac{6\eta'}{r^2\eta} + \frac{24\eta''}{r^3\eta^2} \right) f = 0 \quad (12)$$

where $\eta(r) = \eta_f [1 + \alpha < \chi_1 >_c(r) + \beta < \chi_2 >_c(r)]$ and η' and η'' the first and second order derivatives of η with respect to r . And α and β are directly related to the average stresslets induced by particles of each species.

The effective viscosity can be written in terms of the average stresslets (Batchelor, 1970) as

$$\eta^* = \eta_f (1 + n_1 S_1 + n_2 S_2) = \eta_f (1 + \alpha \phi_1 + \beta \phi_2) \quad (13)$$

where S_j and n_j are average stresslets and number density of particle j , respectively. The limiting value of S_j in very dilute suspensions is $S_{j,0} = 10\pi a_j^3/3$, which directly gives the well-known Einstein's equation for the effective viscosity of very dilute monodisperse suspensions. The average stresslets in Eq. (13) are determined from the average stress acted on the surface of the particle at origin and rate of strain e_{ij} . For example, the average stresslet for extensional flow along the x_1 direction is simply given by

$$S_k = \frac{\langle \sigma_{ij} \rangle}{2 \langle e_{ij} \rangle} = \frac{\langle \sigma_{11} \rangle}{2 \langle e_{11} \rangle} = \frac{2\pi a_k^3}{15} \left[\frac{f''(a_k)}{a_k} - \frac{f'''(a_k)}{3} \right] \quad (14)$$

In the calculations, Eq. (12) is integrated and iteratively solved with initial guess for α and β . We finally determine these coefficients and thus the effective viscosity.

3. Results and discussion

We first compare the theoretical predictions for the effective viscosity of monodisperse suspensions with previous results obtained by experimental or theoretical work. The effective viscosities are plotted as a function of particle volume fraction normalized by the maximum volume fraction in Fig. 3. The results by Brady and Bossis (1985) and Ladd (1990) are based on numerical simulations whereas those by Galadia-Maria (1979) and Bouillot *et al.* (1982) are from experiments. Fig. 3 shows that all the results are in good agreement up to the normalized volume fraction ϕ/ϕ_m of 0.4. Beyond ϕ/ϕ_m of 0.4, the effective viscosities from the present theory are in good agreement with those by Ladd (1990) and Bouillot *et al.* (1982) but are lower than those by Brady and Bossis (1985) and Galadia-Maria (1979). In the paper by Brady and Bossis (1985) it is explained that the increase in the viscosity at high volume fractions results from cluster formation due to lubrication force between the particles near contact from their Stokesian dynamics simulations for effective viscosity of monolayer of monodisperse hard spheres. This simulation condition corresponds to that of experiment by Bouillot *et al.* (1982)

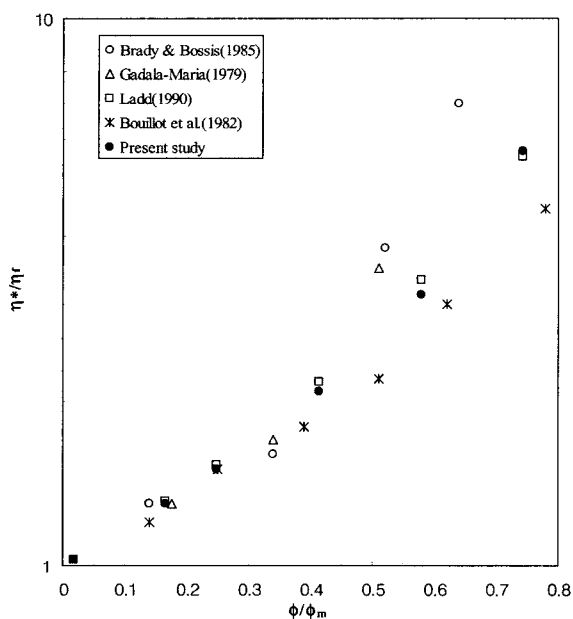


Fig. 3. Effective viscosity of monodisperse suspensions.

whose results for effective viscosity are lower than those by the numerical simulations. However the experimental results by Galadia-Maria (1979) and Pätzold (1980) agree well with this simulation results. It is interesting that the effective viscosities at high volume fractions by Brady and Bossis (1985) are larger than those by Ladd (1990) who has also calculated hydrodynamic interactions among particles including lubrication forces between particles near contact in a similar manner in principle as Brady and Bossis (1985). Ladd's results are, however, in good agreement with those by Sangani and Mo (1988) and Beenakker (1984) based on exact calculations of hydrodynamic interactions among particles. These numerical simulations for computing hydrodynamic interactions among particles use pairwise additivity as an approximation in calculating the multi-particle interactions. In dilute suspensions, the pairwise additivity based on summation of two-particle interaction is sufficient for accurate calculation of multi-particle interactions and three or many particle effects are relatively negligible. Since three or many particle effects become important in dense suspensions, it is likely that numerical simulations underpredict the effective viscosity at high volume fractions of particles due to the limit of pairwise additivity approximation. Another important consideration for high accuracy is inclusion of lubrication force between particles in dense suspensions. Theoretical prediction without lubrication force also results in underestimation of the effective viscosity of dense suspensions.

Fig. 4 shows effective viscosities of bidisperse suspensions as a function of normalized volume fractions ϕ/ϕ_m . Comparison is made among the effective-medium theory modified for bidisperse suspensions, experimental results

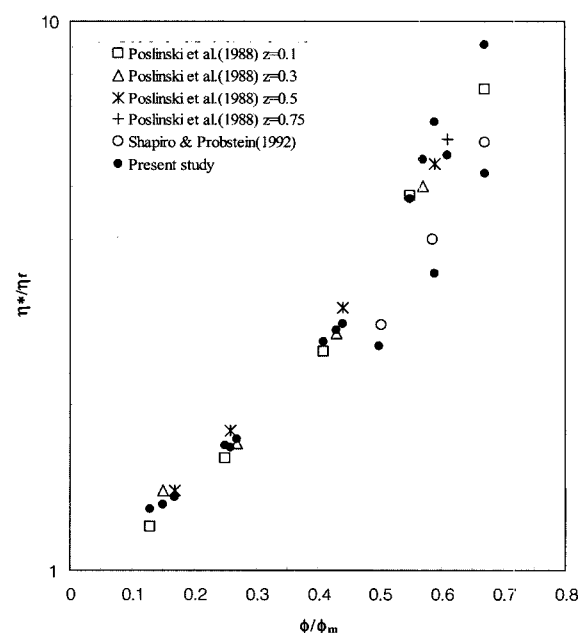


Fig. 4. Effective viscosity of bidisperse suspensions. z is the fraction of small particles to overall volume fraction of particles, ϕ_2/ϕ .

by Poslinski *et al.* (1988) and Shapiro and Probstein (1992), and two dimensional numerical simulations using Stoksonian dynamics by Chang and Powell (1993). Since effective viscosities of bidisperse suspensions are dependent on not only volume fractions but also size ratio and relative volume fraction of each species, calculation of the effective viscosity using modified theory has been done for each condition of experiments. The theoretical predictions are shown to be in excellent agreement with experimental results by Poslinski *et al.* (1988) at each corresponding condition for the whole set of their experiments. It is also seen that there is a small discrepancy between the two beyond ϕ/ϕ_m of 0.6 as seen in monodisperse case. Open circles in Fig. 4 are the experimental results by Shapiro and Probstein (1992) and the theoretical predictions corresponding to these are filled circles below the open circles. The theory gives slightly lower values for the effective viscosity than the experiments at this range of volume fractions. Fig. 4 also shows the results of numerical simulations by Chang and Powell for monolayer of bidisperse suspensions using a numerical method developed by Brady and Bossis (1985). Since unlike monodisperse case the maximum volume fraction of particles is dependent on size ratio and relative volume fraction of each species, the results of two-dimensional simulations are not exactly matched with those of three-dimensional case. These two-dimensional results seem to give slightly higher values than the experimental results by Shapiro and Probstein (1992) for the same size ratio of particles.

4. Conclusions

An effective-medium theory proposed by Acrivos and Chang (1987) was modified to estimate the effective viscosity of suspensions with bidisperse distribution of particle size. Suspensions are modeled as a particle at origin encompassed by an effective-medium in which its properties vary with pair probability. Estimations of the effective viscosity by the modified theory are compared with the results of prior work for bidisperse suspensions as well as for monodisperse case. It is shown that the theoretical estimations in the present study are in good agreement with experimental and other theoretical results up to around 0.45 of the volume fraction normalized by the maximum volume fractions of particles for bidisperse suspensions.

Acknowledgments

This study was supported by research grants from the Korea Science and Engineering Foundation (Project No. R11-2000-088-02009-0) through the Applied Rheology Center.

List of symbols

a	: radius of particle
a_1	: radius of large particle
a_2	: radius of small particle
f	: a function of r
g	: radial distribution function
g_{ij}	: radial distribution function for binary mixture of hard spheres
g_i, \mathbf{g}	: acceleration due to gravity
n_j	: number density of particle species j
p	: pressure
$P(\mathbf{x} \theta)$: probability of finding a particle at \mathbf{x} given a particle at origin
Q_n	: Legendre function
r	: radial distance from the center of the particle at origin
S_j	: stresslet of particle species j
u_i, \mathbf{u}	: velocity of the fluid
\mathbf{x}	: position vector
α, β	: coefficients
χ	: particle phase indicator function
$\langle \chi \rangle_c$: pair probability density, conditional ensemble average of χ
ϕ	: volume fraction of the particles
ϕ_j	: volume fraction of the particle species j
ϕ_m	: maximum volume fraction
η_f	: viscosity of fluid
η^*	: effective viscosity
$\rho(\mathbf{x})$: density at position \mathbf{x}
ρ_f	: density of fluid

ρ_p	: density of particle
σ_{ij}	: stress tensor
ψ	: stream function

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Appendix: Radial distribution function of binary mixture of hard spheres

We followed the calculation procedure for $g_{ij}(r)$ given by Throop and Bearman (1965). The inversion integrals of the Laplace transforms for $g_{ij}(r)$ are given by

$$rg_{11}(r) = \frac{1}{12\xi_1} \sum_{m=0}^{\infty} \frac{1}{2\pi i} \int \frac{s[H-L_2(s)\exp(2sa_2)][I(s)]^m \exp[s(r-2a_1-2a_2)]ds}{[F(s)]^{m+1}}$$

$$rg_{22}(r) = \frac{1}{12\xi_2} \sum_{m=0}^{\infty} \frac{1}{2\pi i} \int \frac{s[H-L_1(s)\exp(2sa_1)][I(s)]^m \exp[s(r-2a_1-2a_2)]ds}{[F(s)]^{m+1}}$$

$$rg_{12}(r) = \sum_{m=0}^{\infty} \frac{1}{2\pi i} \int \left[12(\xi_2 a_2^3 - \xi_1 a_1^3)(a_2 - a_1) - a_{12} \left(1 - \frac{1}{2}\phi\right) \right] s \frac{[I(s)]^m s^2 \exp[s(r-a_{12})]ds}{[F(s)]^{m+1}} \quad (A1)$$

where $\xi = \pi n_i/6$, n_i being the number density of species i . H , $L_1(s)$, $L_2(s)$, $F(s)$ and $I(s)$ are given by

$$H = 72\xi_1\xi_2(a_2 - a_1)$$

$$L_1(s) = 12\xi_2 \left[\left(1 + \frac{1}{2}\phi\right) + 12\xi_1 a_1^2 (a_2 - a_1) \right] a_2 s^2 + [12\xi_2(1+2\phi) - 2Ha_1]s + H$$

$$L_2(s) = 12\xi_1 \left[\left(1 + \frac{1}{2}\phi\right) + 12\xi_2 a_2^2 (a_1 - a_2) \right] a_1 s^2 + [12\xi_1(1+2\phi) - 2Ha_2]s + H$$

$$F(s) = H + [12(\xi_1 + \xi_2)(1+2\phi) - 2H(a_1 + a_2)]s - 72(\xi_1 a_1^2 + \xi_2 a_2^2)s^2 - 24(\xi_1 a_1^2 + \xi_2 a_2^2)(1-\phi)s^3 - (1-\phi)^2 s^4$$

$$I(s) = L_2(s)\exp(-2sa_1) + L_1(s)\exp(-2sa_2) - H\exp[-2s(a_1 + a_2)]. \quad (A2)$$

Using the residual theorem the integrals in Eq. (A2) equal to $2\pi i R_m^{ij}$ where

$$R_m^{ij} = \frac{1}{(m-1)!} \sum_{i=1}^4 \lim_{s \rightarrow t_i} \left(\frac{d^{m-1}}{ds^{m-1}} [(s-t_i)^m] \sum_{k=1}^2 \frac{\phi_k^{ij}(s) \exp[s(r-\psi_k^{ij}(a_1, a_2))]}{[F(s)]^m} \right) \quad (A3)$$

where t_i correspond to the four roots of $F(s) = 0$. Here, ϕ_k^{ij} are polynomials in s and are given by

$$\phi_1^{11} = \frac{-L_2(s)s}{12\xi_1} \Xi(s), \quad \phi_2^{11} = \frac{Hs}{12\xi_1} \Xi(s)$$

$$\phi_1^{22} = \frac{-L_1(s)s}{12\xi_2} \Xi(s), \quad \phi_2^{22} = \frac{Hs}{12\xi_2} \Xi(s)$$

$$\phi_1^{12} = \left\{ \left[12(\xi_2 a_2^3 - \xi_1 a_1^3)(a_2 - a_1) - a_{12} \left(1 - \frac{\phi}{2}\right) \right] s - (1+2\phi) \right\} s^2 \Xi(s),$$

$$\phi_2^{12} = 0 \quad (A4)$$

where

$$\Xi(s) = \sum_{q_1=0}^{q_1+} \sum_{q_2=0}^{q_2+} \sum_{q_3=0}^{q_3=m-1} \frac{(m-1)!}{q_1!q_2!q_3!} L(s)^{q_1} L(s)^{q_2} (-H)^{q_3} \quad (A5)$$

ψ_k^{ij} are linear combinations of a_1 and a_2

$$\psi_1^{11} = 2(m-q_2)a_1 + 2(m-q_1-1)a_2$$

$$\psi_2^{11} = 2(m-q_2)a_1 + 2(m-q_1)a_2$$

$$\psi_1^{22} = 2(m-q_2-1)a_1 + 2(m-q_1-1)a_2$$

$$\psi_2^{22} = 2(m-q_2)a_1 + 2(m-q_1)a_2$$

$$\psi_1^{12} = 2\left(m-q_2-\frac{1}{2}\right)a_1 + 2\left(m-q_1-\frac{1}{2}\right)a_2$$

$$\psi_2^{12} = 0 \quad (A6)$$

Now $g_{ij}(r)$ can be determined by carrying out the differentiation in Eq. (A4). Since the contour integrals in Eq. (A1) equal the sum of residues R_m^{ij} for $r - \psi_k^{ij}(a_1, a_2) > 0$ and are zero otherwise, the evaluation of g_{ij} as a function of r is limited by the differentiation order m . Calculation of $g_{ij}(r)$ at large r requires higher order differentiation which becomes quite cumbersome. In the present study, the differentiation as carried out up to $m = 4$ which is sufficient to determine g_{ij} for $r < 8a_1 + 2a_2$. Beyond this distance, $g_{ij}(r)$ was taken to be unity in our calculations.