Complex viscosity of dilute capsule suspensions: 
a numerical study

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Abstract

In this paper, we apply an oscillating shear flow to a dilute capsule suspension and report its viscoelastic properties. We analyze the complex viscosity under different capillary numbers and viscosity ratios, which is a viscosity contrast inside and outside the capsules. For all viscosity ratios, the real part of complex viscosity $\eta'$ monotonically decreases with the frequency of the applied oscillating shear, while the imaginary part $\eta''$ shows the maximum value at an intermediate frequency. In general, the capsule with a larger viscosity ratio gives larger $\eta'$, while that of smaller viscosity ratio gives larger $\eta''$. At high frequencies, the capsule that has higher (lower) inner viscosity contributes to increase (decrease) the viscosity of the solutions. In order to separately discuss the contributions of the membrane elasticity and internal fluid viscosity, we analyse the first term and second term of the particle stress tensor. The first term, which is called elastic stress in this paper, represents particle stress that arises from the capsule deformation. The amplitude of elastic stress is nearly constant at low frequencies, while it is inversely proportional to the applied frequency at high frequencies. The phase of elastic stress shifts from the shear to strain phases when the frequency increases. These tendencies of elastic stress do not depend on the viscosity ratio, and the qualitative trends are the same for all viscosity ratios. The second term, which is called viscous stress in this paper, represents particle stress that arises from the viscosity ratio, and the trend is drastically different by the viscosity ratio. The viscous stress contributes to increase (decrease) the viscosity and decrease (increase) the elasticity, when the capsule inner viscosity is higher (lower). Finally, we evaluate the effect of the capillary number. At low frequencies, both the capillary number and viscosity ratio are important factors for the rheology. On the other hand, the viscosity ratio becomes the only governing factor at high frequencies because the membrane elasticity has a negligible effect.

Keywords : Capsule, Rheology, Complex viscosity, Stokes flow, Boundary element method

1. Introduction

A capsule is a liquid drop enclosed by a deformable membrane. Capsules are found in both nature and artificial products, such as biological cells and membrane-bounded drugs. Since it is important to understand how the presence of capsules would impact the suspension rheology, there are many works both in dilute (for example: Ramanujan and Pozrikidis 1998; Clausen and Aidun 2010; Foess et al. 2011) and dense (Clausen et al. 2011; Matsunaga et al. 2015; Rosti et al. 2018) capsule suspensions under simple shear flow. These previous studies can be categorized into the research field of biorheology because blood contains many red blood cells, which can be modelled as the capsules.
In recent years, several researchers started to investigate the dynamics of capsules in unsteady flow (Zhao and Bagchi 2011; Matsunaga et al. 2015; Zhu et al. 2015; Cordasco and Bagchi 2016). In our previous study (Matsunaga et al. 2015), we reported the capsule deformation under oscillating shear flow. We found that the capsule deformation is large and the phase of the capsule deformation is nearly the same as phase of the applied shear, when the frequency of the applied shear is low. By increasing the frequency, the capsule deformation starts to decay with a scale inversely proportional to the frequency. Together with the deformation decay, the deformation phase gradually shifts toward the strain phase. We also proposed an equation to estimate the threshold frequency that the deformation decay starts.

Despite the understanding of the capsule behaviour under oscillating shear, the suspension rheology such as the viscoelasticity is not well understood. There is an analytical study on the complex viscosity of a shell suspension (Sakanishi and Ferry 1983), but the knowledge is limited to small deformations. In this paper, we evaluate the complex viscosity of dilute suspensions that contain a capsule with different viscosity ratios. Note that the viscosity ratio is a viscosity contrast between inside and outside the capsule. Our goal is to clarify how the rheology changes with the viscosity ratio and capillary number.

2. Governing equations and Methods

Consider a spherical capsule with radius $a$, suspended in an unbounded flow field with viscosity $\mu$ and density $\rho$ as shown in Fig. 1. The fluid inside the capsule has viscosity $\lambda \mu$ and density $\rho$, where $\lambda$ is the viscosity ratio. The capsule membrane is modelled as a hyperelastic sheet with negligible thickness. The volume fraction of the capsule $\phi = 4\pi a^3/3V$ ($V$ is the control volume) is assumed to be $\phi \ll 1$ ($V \to \infty$), and the capsule-capsule interactions are neglected.

The oscillating shear flow is given by

$$\dot{\gamma}(t) = \dot{\gamma}_0 \exp j(2\pi ft),$$  \hspace{1cm} (1)

where $\dot{\gamma}_0$ is the oscillation amplitude of the shear rate, $j$ is the imaginary unit and $f$ is the oscillation frequency (see also Fig. 2). The corresponding strain $\gamma(t)$ is then

$$\gamma(t) = \int \dot{\gamma}(t)dt = \frac{\dot{\gamma}_0}{2\pi f} \exp j(2\pi ft),$$  \hspace{1cm} (2)

where the strain amplitude $\gamma_0$ is

$$\gamma_0 = \frac{\dot{\gamma}_0}{2\pi f}.$$  \hspace{1cm} (3)

Note that the phase of the strain lags behind the shear rate by $\pi/2$. The particle Reynolds number $Re_p = \rho \dot{\gamma}_0 a^2/\mu$ is assumed to be small enough to treat the velocity field as a Stokes flow. The frequency parameter, $\beta = \rho a^2 f/\mu$ (Pozrikidis 1992), is also assumed to be small enough to disregard of the effect of flow unsteadiness. For instance, suppose that a capsule with radius $a = 1.0 \times 10^{-5}$ m suspended in a fluid with shear rate $\dot{\gamma}_0 = 10.0$ s$^{-1}$, density $\rho = 1.0 \times 10^3$ kg/m$^3$, and viscosity $\mu = 1.0 \times 10^{-3}$ Pa$\cdot$s. Under these conditions, the applicable range
of the frequency is \( f = 10^{-1} - 10^3 \text{ s}^{-1} \) \((f/\gamma_0 = 10^{-2} - 10^2)\), which corresponds to a particle Reynolds number \( \text{Re}_p = 1.0 \times 10^3 \) and a frequency parameter \( \beta = 10^{-5} - 10^{-1} \). Note that the parameter range includes the condition of microcirculations \((f/\gamma_0 = 10^{-2}; f = 1 \text{ s}^{-1}, \gamma_0 = 100 \text{ s}^{-1})\) of our body.

2.1. Fluid mechanics

The Stokes flow velocity of a given observation point \( x \) is described by the boundary integral formulation (Pozrikidis 1992; Foessel et al. 2011):

\[
\mathbf{v}(x) = \mathbf{v}^{\infty}(x) - \frac{1}{8\pi\mu} \int_A \mathbf{J}(x,y) \cdot \mathbf{q}(y) dA(y) + \frac{1 - A}{8\pi} \int_A \mathbf{v}(y) \cdot \mathbf{K}(x,y) \cdot n(y) dA(y),
\]

where \( \mathbf{n} \) is the outward unit normal vector at the membrane material point \( y \), \( A \) is the capsule surface and \( \mathbf{q}(y) \) is the traction jump across the membrane, which can be also described as

\[
\mathbf{q}(y) = [\mathbf{\sigma}_{\text{out}}(y) - \mathbf{\sigma}_{\text{in}}(y)] \cdot \mathbf{n}(y)
\]

where \( \mathbf{\sigma}_{\text{in}} \) and \( \mathbf{\sigma}_{\text{out}} \) are the stress tensors on the inner and outer surfaces of the capsule membrane, respectively. Using Eq. (1), the background flow field \( \mathbf{v}^{\infty} \) is given by

\[
(e^{\infty}_1, e^{\infty}_2, e^{\infty}_3) = (\gamma(t)x_3, 0, 0).
\]

Note that indices 1-3 correspond to the directions in the Cartesian coordinate. The 2nd-order tensor \( \mathbf{J} \) and the 3rd-order tensor \( \mathbf{K} \) are the Green’s function of single- and double-layer potentials, which are defined by

\[
\mathbf{J} = \frac{1}{r} + \frac{r \otimes r}{r^3},
\]

\[
\mathbf{K} = 6 \frac{r \otimes r \otimes r}{r^5},
\]

where \( r = x - y \), \( r = |r| \) and \( \mathbf{I} \) is the identity tensor.

2.2. Membrane mechanics

The membrane is modelled as an isotropic and hyperelastic material that follows the neo-Hookean constitutive law. The bending rigidity is assumed to be negligible because of the small thickness, and only the in-plane stress is considered. The surface deformation gradient tensor \( \mathbf{F}_s \) is given by

\[
dx = \mathbf{F}_s \cdot dX,
\]

where \( \mathbf{X} \) and \( x \) are the membrane material points of the reference and deformed states, respectively. The Cauchy stress tensor \( \mathbf{T} \) is

\[
\mathbf{T} = \frac{1}{J_s} \mathbf{F}_s \cdot \frac{\partial \mathbf{w}_s}{\partial \mathbf{e}} \cdot \mathbf{F}_s^T,
\]

where \( J_s = \lambda_1 \lambda_2 \) represents the area dilation ratio and \( \mathbf{e} \) is the Green-Lagrange strain tensor that is given by

\[
\mathbf{e} = \frac{1}{2} \left( \mathbf{F}_s^T \cdot \mathbf{F}_s - \mathbf{I}_s \right).
\]

Note that \( \mathbf{I}_s \) is the tangential projection operator. The strain energy function \( w_s \) of the neo-Hookean constitutive law (Pozrikidis 2003) is

\[
w_s = \frac{G_s}{2} \left( I_1 - 1 + \frac{1}{I_2 + 2} \right),
\]

where \( G_s \) is the surface shear elastic modulus and \( I_1, I_2 \) are invariants defined as \( I_1 = 2\text{tr}(\mathbf{e}) \) and \( I_2 = 2\text{det}(\mathbf{e}) \).
From the definition above, a conversion from $\eta$ to $\eta^*$ is given as
\begin{equation}
\eta^* = \frac{\tau_0}{\gamma_0}j\frac{2\pi f}{\gamma_0}, \quad \eta' = \frac{\tau_0}{\gamma_0}\cos(\delta_\tau), \quad \eta'' = -\frac{\tau_0}{\gamma_0}\sin(\delta_\tau).
\end{equation}

Using the amplitude of the complex viscosity
\begin{equation}
|\eta^*| = \sqrt{(\eta')^2 + (\eta'')^2} = \frac{\tau_0}{\gamma_0},
\end{equation}

$\eta'$ and $\eta''$ can be rewritten as follows,
\begin{equation}
\eta' = |\eta^*|\cos(\delta_\tau), \quad \eta'' = -|\eta^*|\sin(\delta_\tau).
\end{equation}

From the definition above, a conversion from $(G', \eta^*)$ to $(\eta', \eta'')$ is given as
\begin{equation}
\eta' = \frac{1}{2\pi f}G'', \quad \eta'' = \frac{1}{2\pi f}G'.
\end{equation}
In this paper, we use \((\eta', \eta'')\) to compare with previous studies (Sakanishi and Ferry 1983; Farutin and Misbah 2012). Shear stress due to the capsule existence is calculated using the particle stress tensor \(\Sigma_{ij}^{(p)}\) (Batchelor 1970):

\[
\Sigma_{ij}^{(p)} = \frac{1}{V} \int_A \left\{ \frac{1}{2} (x_i q_j + q_i x_j) - \mu(1 - \lambda)(\epsilon_i n_j + n_i \epsilon_j) \right\} dA
\]

(25)

where indices \(i, j\) are in a range 1-3. First term of equation is a stress due to the membrane elasticity, and the second term is a stress due to the viscosity ratio \(\lambda\). Note that all the components of \(\Sigma_{ij}^{(p)}\) are zero at the resting state, and they would have non-zero values under the deformation \(q \neq 0\) or movement \(v \neq 0\).

2.4. Numerical method

In this study, we use the boundary element method for fluid mechanics and the finite element method for membrane mechanics (Walter et al. 2010; Foessel et al. 2011). Note we used these numerical methods in our previous studies (Matsunaga et al. 2014 2015 2016a 2016b; Imai and Matsunaga 2017; Ito et al. 2019), and see these works for the detailed implementation.

An unstructured triangular mesh with 2,562 nodes and 5,120 linear elements is used to discretize the capsule membrane. In the boundary element method, the Gaussian quadrature method is used to compute the integral over triangular elements. For singular elements, a polar coordinate is introduced to get rid of the \(1/r\) singularity (Pozrikidis 1995). The weak form of the equilibrium equation on the capsule,

\[
\int_A \hat{u} \cdot q dA = \int_A \hat{\epsilon} : T dA,
\]

(26)

is solved by the finite element method, where \(\hat{u}\) is the virtual displacement and \(\hat{\epsilon}\) is the virtual strain. The membrane velocity is given by the kinematic condition:

\[
\frac{dx}{dt} = v(x).
\]

(27)

An explicit second-order Runge-Kutta method is used to update the position of the membrane material points.

In order to determine the shear stress amplitude \(\tau_0\) and the phase difference of the stress \(\delta\), we apply DFT (discrete Fourier transform) to \(\Sigma_{ij}^{(p)}\), which is the shear component of the particle stress tensor. In this study, we show the complex viscosity that is normalized by the volume fraction \(\phi\) and viscosity \(\mu\) as

\[
\frac{\eta'}{\mu \phi} = \frac{\tau_0}{\mu \gamma_0 \phi} \exp j \delta, \quad \text{(28)}
\]

and

\[
\frac{\eta''}{\mu \phi} = + \frac{\tau_0}{\mu \gamma_0 \phi} \cos(\delta), \quad \text{(29)}
\]

\[
\frac{\eta'''}{\mu \phi} = - \frac{\tau_0}{\mu \gamma_0 \phi} \sin(\delta). \quad \text{(30)}
\]

2.4.1. Dimensionless parameters

Three non-dimensional parameters are considered: the capillary number \(Ca\), the non-dimensional frequency \(f/\gamma_0\) and the viscosity ratio \(\lambda\). The capillary number,

\[
Ca = \frac{\mu \gamma_0 a}{G}, \quad \text{(31)}
\]

describes the strength of the viscous force relative to the elastic force of the membrane. The non-dimensional frequency, which is similar to the Strouhal number, describes the frequency of the oscillation compared to the strength of the shear rate.

3. Results

Before presenting the rheological properties, we summarize the capsule deformation under oscillating shear flow (Matsunaga et al., 2015). When the frequency of applied shear \(f/\gamma_0\) is low, the capsule deformation is large and the phase of the capsule deformation \(\delta_D\) is nearly the same as the applied shear \(\delta_D = 0\). Increasing
the frequency, the capsule deformation starts to decay with $f^{-1}$ around a threshold frequency $(f/\dot{\gamma}_0)_{th}$. Together with the deformation decay, the deformation phase gradually shifts toward the strain phase $\delta_D = -\pi/2$. The threshold frequency can be predicted using a relation

$$
(f/\dot{\gamma}_0)_{th} = \frac{5}{4\pi D_{12}^{sd}(2\lambda + 3)},
$$

(32)

where $D_{12}^{sd}$ is the Taylor deformation of capsules under steady shear flow. In this section, we first present the viscoelasticity of a solution that has a capsule with the viscosity ratio $\lambda = 1$. Secondly, we discuss the effect of the viscosity ratio $\lambda$. Finally, we report the effect of the capillary number $Ca$.

### 3.1. Dilute capsule suspension with viscosity ratio $\lambda = 1$

First, we investigate the complex viscosity of a solution with a capsule $\lambda = 1$. In this analysis, we can focus on the first term of the particle stress tensor (25) since the second term is omitted for $\lambda = 1$. Figure 3 shows (a) the shear stress amplitude $\tau_0/(\mu \dot{\gamma}_0 \phi)$ and (b) its phase difference $\delta$ from the phase of shear rate. Considering that the larger capsule deformation produces larger shear stress $\Sigma^{(p)}$, the deformation amplitude and phase are directly related the shear stress $\tau_0$. Same as the deformation decay (Matsunaga et al. 2015), the stress amplitude also decay with $f^{-1}$ after a threshold frequency $(f/\dot{\gamma}_0)_{th} \approx 0.16$. Figure 3(b) shows that the phase of particle stress $\delta$ also shift from the 0 (phase of shear rate $\dot{\gamma}(t)$) to $-\pi/2$ (phase of strain $\gamma(t)$) same as the deformation phase $\delta_D$ (Matsunaga et al. 2015).

Figure 3(c) shows the complex viscosity $\eta'$ and $\eta''$ evaluated from the stress amplitude $\tau_0/(\mu \dot{\gamma}_0 \phi)$ and the phase difference $\delta$. The figure shows that the $\eta'$ monotonically decreases with the frequency $f$, while $\eta''$ shows a peak at an intermediate frequency. This tendency is in good agreement with the analytical models of emulsions.
(Oldroyd 1953) and spherical shells (Sakanishi and Ferry, 1983), and the simulation of vesicles (Farutin and Misbah, 2012). The decrease in $\eta'$ and the increase in $\eta''$ at low frequencies ($f/\gamma_0 < (f/\gamma_0)_{ba}$) can be explained by the phase shift, because the phase shift toward $\delta = -\pi/2$ occurs while the stress amplitude is nearly constant. Since the phase shift toward $\delta = -\pi/2$ occurs together with the decay of the stress amplitude at high frequencies ($f/\gamma_0 > (f/\gamma_0)_{ba}$), a decrease in $\eta'$ is caused by both the phase shift and the decrease of the stress amplitude. Also, a decrease in $\eta''$ can be explained by the decrease of the stress amplitude.

### 3.2. Effect of the viscosity ratio

Next, we discuss the effect of viscosity ratio $\lambda$. Figure 4(c) shows the dynamic viscosity coefficient $\eta'$ drastically differs by the viscosity ratios. The value $\eta'$ always shows positive values with a small dependence on the frequency for $\lambda > 1$, while it goes to negative values by increasing the frequency for $\lambda < 1$. This result indicates that the capsule with $\lambda > 1$ contributes to increase the suspension viscosity at any frequencies, while the capsule with $\lambda < 1$ contributes to decrease the viscosity at high frequencies. This tendency is intuitive to understand because the capsules with higher inner viscosity $\lambda \mu$ exhibit larger viscosity increase. Figure 4(d) shows that $\eta''$ has the maximum value at the non-dimensional frequency $f/\gamma_0 = 2.0 \times 10^{-1}$, and $\eta''$ is larger for lower viscosity ratios $\lambda$. This result has an agreement with the analysis of the shell model (Sakanishi and Ferry 1983) and vesicles (Farutin and Misbah 2012). Contrary to the capsule with $\lambda = 1$, the stress amplitude converges to certain values for $\lambda \neq 1$ as shown in Fig. 4(a). Figure 4(b) shows that the stress phase $\delta$ also drastically changes by the viscosity ratio. Capsules with $\lambda > 1$ give particle stress that has the same phase as the applied shear $\delta = 0$, while capsules with $\lambda < 1$ gives the opposite phase $\delta = -\pi$ at high frequencies.

We plot the results of $\eta''$ in a complex plane, with $\eta''/\mu \phi$ on the horizontal axis and $\eta''/\mu \phi$ on the vertical axis, in Fig. 5. When the frequency increases, the plots move from right to left in a semicircular orbit for all cases, and the size of the semicircle gets smaller by increasing the viscosity ratio $\lambda$. The figure suggests that the complex viscosity has a weak dependence of the viscosity ratio $\lambda$ at low frequencies, while it causes a large
The complex viscosity $\eta^*$ for capsule with different viscosity ratios, under capillary number $Ca = 0.5$.

difference at higher frequencies. By further increasing the viscosity ratio, we confirm that the complex viscosity converges to $(\eta'/\mu \phi, \eta''/\mu \phi) = (2.5, 0.0)$ (data not shown), which corresponds to the rheology of diluted rigid sphere suspension (Einstein1906).

For a detailed analysis, we divide the particle stress tensor (25) into two terms: the elastic $\{\Sigma^{(p)} \}_{i,j}$ and viscosity ratio term $\{\Sigma^{(p)} \}_{i,j}^\lambda$, which is defined as

$$\Sigma^{(p)}_{i,j} = \{\Sigma^{(p)} \}_{i,j}^e + \{\Sigma^{(p)} \}_{i,j}^\lambda \quad \text{(33)}$$

$$\{\Sigma^{(p)} \}_{i,j}^e = \frac{1}{V} \int_A \left\{ \frac{1}{2} (x_i q_j + q_i x_j) \right\} dA, \quad \text{(34)}$$

$$\{\Sigma^{(p)} \}_{i,j}^\lambda = -\frac{1}{V} \int_A \left\{ \mu (1-\lambda) (n_i n_j + n_j n_i) \right\} dA. \quad \text{(35)}$$

For convenience, we call $\{\Sigma^{(p)} \}_{i,j}^e$ and $\{\Sigma^{(p)} \}_{i,j}^\lambda$ as elastic stress and viscous stress respectively in this paper. The stress amplitude $\tau_\epsilon$ and the phase difference $\delta_\epsilon$ are obtained by applying DFT to the shear stress $\{\Sigma^{(p)} \}_{i,j}^e$, and the elastic term contribution to the complex viscosity is given by

$$\eta^*_e = \frac{\tau_\epsilon}{\mu \dot{\gamma} \phi} \exp j \delta_\epsilon = \eta'_e - j \eta''_e. \quad \text{(36)}$$

Similarly, the viscous term contribution to the complex viscosity is given by

$$\eta^*_\lambda = \frac{\tau_\lambda}{\mu \dot{\gamma} \phi} \exp j \delta_\lambda = \eta'_\lambda - j \eta''_\lambda. \quad \text{(37)}$$

The complex viscosity $\eta^*_e$ and $\eta^*_\lambda$ satisfies following relations,

$$\eta' = \eta'_e + \eta'_\lambda, \quad \text{(38)}$$

$$\eta'' = \eta''_e + \eta''_\lambda, \quad \text{(39)}$$

because of the linearity of the system.

### 3.2.1. Elastic stress

Figure 6 shows contribution of the elastic stress $\{\Sigma^{(p)} \}_{i,j}^e$. As we discussed in the previous sections, the capsule deformation and its phase are directly related to the shear stress. Recalling that the capsule has a larger deformation for lower viscosity ratio $\lambda$ (Ramanujan and Pozrikidis 1998; Foessel et al. 2011), the stress amplitude is larger for lower $\lambda$ as shown in Fig. 6(a). Similar to the case of $\lambda = 1$, the phase $\delta_\epsilon$ monotonously decreases with the frequency and converges to the phase of strain, $-\pi/2$. This phase shift is in good agreement.
with the phase shift of the deformation phase $\delta_D$, which is reported in our previous work (Matsunaga et al. 2015).

Figures 6(c) and (d) show the elastic stress contribution of the complex viscosity, $\eta'_e$ and $\eta''_e$. The figure indicates that lower viscosity ratio $\lambda$ leads to higher $\eta'_e$ and $\eta''_e$, which is based on the larger elastic stress $\tau_e$. Comparing the curves with that of $\lambda = 1$, we found that qualitative trends of both $\eta'_e$ and $\eta''_e$ do not change by the viscosity ratio. Therefore, the viscous stress $(\Sigma_{12}^{(p)})_1$ must be the governing factor to characterize the complex viscosity.

3.2.2. Viscous stress

Figure 7 shows contribution of the viscous stress $(\Sigma_{12}^{(p)})_1$. Two figures, Fig. 7(c) and (d), show that the signs of both $\eta'_e$ and $\eta''_e$ are different between $\lambda > 1$ and $< 1$. The value $\eta'_e$ is positive for $\lambda > 1$ but is negative for $\lambda < 1$, while this trend is opposite for $\eta''_e$. These indicate that the capsule with viscosity ratio $\lambda > 1$ ($\lambda < 1$) contributes to increase (decrease) the viscosity, and decrease (increase) the elasticity of the solutions.

Since the stress amplitude $\tau_1$ increases with the frequency $f$ while $\tau_e$ decreases, the viscous stress $(\Sigma_{12}^{(p)})_1$ becomes a dominant factor at high frequencies. Contrary to the phase $\delta_e$, $\delta_1$ is nearly constant throughout the frequencies and the phases stay $\approx 0$ ($\lambda > 1$) or $\approx -\pi$ ($\lambda < 1$) depending on the viscosity ratio. Since there is no significant change in the stress amplitude $\tau_1$, we can conclude that the phase of viscous stress characterise the signs of $\eta'$ and $\eta''$.

3.3. Effect of the capillary number

Finally, we clarify the effect of the capillary number in this final subsection. Note that the capillary number was all fixed to $Ca = 0.5$ in the previous sections. Figure 8 shows the complex viscosity $\eta'$ for different capillary number $Ca$ and viscosity ratio $\lambda$. Same as Fig. 5, all the plots move from right to left in semicircular orbits with increasing $f$.

Figure 8 shows that the capillary number $Ca$ does not affect the qualitative tendency of the complex viscosity.
viscosity, and it only changes the stress amplitude at low frequencies. The stress amplitude \( \tau = |\eta'\eta''| \) is larger for a lower capillary number \( Ca \), and this shear thinning character is also observed under simple shear flow (Ramanujan and Pozrikidis 1998; Baguchi and Kalluri 2010; Clausen and Aidun 2010). The plots start to converge to a same orbit at the threshold frequency \((f/\dot{\gamma}_0)_{th}\), and this is because the membrane elasticity have negligible effect to the suspension rheology at high frequencies \(f/\dot{\gamma}_0 > (f/\dot{\gamma}_0)_{th}\).

4. Conclusion

In this paper, we present the complex viscosity of the dilute capsule suspensions. For all viscosity ratios, the real part of complex viscosity \( \eta' \) monotonically decreases with the frequency, while the imaginary part \( \eta'' \) shows the maximum value at an intermediate frequency. In general, the capsule with a larger viscosity ratio gives larger \( \eta' \), while that of smaller viscosity ratio gives larger \( \eta'' \). At high frequencies, the capsule that has higher (lower) inner viscosity contributes to increase (decrease) the viscosity of the solutions. These qualitative tendencies match with the previous studies on the elastic shell and vesicle models.

In order to separately discuss the contributions of the membrane elasticity and internal fluid viscosity, we analyzed the first term and second term of the particle stress tensor. The first term, which is called elastic stress in this paper, represents particle stress that arises from the capsule deformation (Matsunaga et al. 2015). The amplitude of elastic stress is nearly constant at low frequencies, while it is inversely proportional to the applied frequency at high frequencies. The phase of elastic stress shifts from the shear to strain phases when the frequency increases. These tendencies of elastic stress do not depend on the viscosity ratio, and the qualitative trends are the same for all viscosity ratios. The second term, which is called viscous stress in this paper, represents particle stress that arises from the viscosity ratio, and the trend is drastically different by the viscosity ratio. The viscous stress contributes to increase (decrease) the viscosity and decrease (increase) the elasticity, when the capsule inner viscosity is higher (lower). Finally, we evaluate the effect of the capillary number. At low frequencies, both the capillary number and viscosity ratio are important factors for the rheology. On the
The complex viscosity $\eta^*$ for capsule with different viscosity ratios: (a) $\lambda = 0.5$, (b) 1.0 and (c) 5.0.

other hand, the viscosity ratio becomes the only governing factor at high frequencies because the membrane elasticity has a negligible effect. For the future works, we also summarize here the limitations of our approach and unsolved problems. The first limitation is the applicable range of the Reynolds number. Our numerical method is limited to small Reynolds number regime (Stokes flow) and cannot evaluate the effect of inertia. It would be interesting to discuss the effect of inertia in the future works. The second limitation is the confinement effect. We assume that there is no wall confinement in this study, but it will be interesting to clarify how the wall confinement (such as the cone-plate rheometer) would modify the suspension rheology.

Compared to many finding of the capsule suspension rheology under steady shear flow, its rheology under oscillating shear flow has been limited. We believe that our finding would be an essential building block for the future finding in the capsule suspension rheology, and also in the research field of biorheology.

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