Numerical Simulation of Mass-Transfer Characteristics of a Bubble Rising in Yield Stress Fluids

Baixu Cao, Jungeng Fan, Xuelin Sun, and Shaobai Li*

ABSTRACT: The mass-transfer characteristics of bubbles rising in yield stress fluids was investigated numerically using volume-of-fluid and user-defined function methods in this study. The CO₂ concentration profiles inside the liquid phase near the bubble equator at different bubble diameters, yield stresses, consistency coefficients, and flow indices were observed. The results revealed that the rate of mass transfer decreased with the increase of yield stress, consistency coefficient, and flow index of the liquid phase and the decrease of bubble diameter. Moreover, two empirical correlations for the drag coefficients and Sherwood numbers were developed by introducing one correction factor X for C_D correlation and another correction factor f_s for Sherwood numbers for correcting the influence of yield stress behavior on bubble motion and mass transfer, respectively. The predictions of the two correlations were compared with the simulated data, and a satisfactory agreement was found.

INTRODUCTION

The phenomenon of bubble motion in the liquid phase occurs in many industrial processes, including chemical engineering, fermentation, metallurgy, energy, and environment.1 As we know, these processes would involve the chemical reactions between the components of gas and liquid phases. In addition, the mass-transfer behavior could control the chemical reactions between the gas and liquid phases in the above-mentioned process.2 Therefore, many researchers attempted to study the mass-transfer behavior in gas–liquid phases experimentally. The focus of these investigations was primarily on the influencing factor of mass transfer, such as the gas nozzle configuration,3 gas flow rate,4 pH value of the liquid phase,5 bubble column diameter,6 liquid properties,7–9 and so on.

In comparison to the gas–liquid mass-transfer process in Newtonian fluids, much less is known about the gas–liquid mass-transfer behaviors in non-Newtonian fluids, which is highly prevailing in a variety of biological processes such as biochemical fermentation, cell culture, biological wastewater treatment, and so on. Owing to complex rheological properties, the mass-transfer process from bubbles to the liquid phase remains in an elementary stage. However, a profound understanding of mass transfer from bubbles to such liquid phase media is a precondition to designing and operating efficiently in substantial industrial processes.10 In the last few years, there were several works about the mass transport process from bubbles to the non-Newtonian liquid. Gómez-Díaz et al.11,12 investigated the effects of rheological properties and gas flow rate on the mass-transfer rate. Park et al.13,14 studied the mass transfer of CO₂ to non-Newtonian fluids accompanied by chemical reactions. Kilonzo et al.15 reviewed the influencing factors of oxygen mass transfer in non-Newtonian fluids of gas-lift bioreactors. So far, most works are about collecting macroscopic information, and few studies on the microcosmic mechanism of the mass transfer surrounding the bubble are available. The microcosmic mechanism of the mass transfer is the basis for designing gas–liquid reactors; in addition, for a number of biochemical processes, local forces such as shear and yield stresses may cause damage to microorganisms or cells.16 Thus, it is greatly significant to understand the gas–liquid mass-transfer mechanism occurring on a microscale. Nevertheless, it is extremely difficult to obtain the flow field and concentration field around a bubble on a microscopic view via traditional experimental methods. In fact, the numerical simulation method has become a powerful tool for attaining the details of the flow structure and the mass-transfer mechanism around the bubble. For several years, several attempts have been made to understand the microscale flow and mass-transfer behavior of bubbles. Koynov et al.17 observed mass transfer and chemical reaction of bubble swarms and a single bubble in the liquid phase and found that the mass transfer of bubble swarms differs from that of the single bubble. Bothe et al.18 examined

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the transfer and transport of oxygen from a single bubble in aqueous solutions using the advanced volume-of-fluid (VOF) method. Radl et al. investigated the motion and mass transfer of deformable bubbles in non-Newtonian fluids using a 2-D numerical simulation method, and the non-Newtonian fluids reveal viscoelastic and shear thinning properties.

Yield stress fluids are a profoundly crucial class of non-Newtonian fluids, which behave like a solid when the applied shear stress is below the yield stress; nonetheless, they will flow like fluids when the applied shear stress exceeds the yield stress. The motion behavior of bubbles in yield stress fluids is imperative in many practical applications, such as in food processing, waste processing, daily chemical industry, oil extraction, and biochemical reactors. Accordingly, this topic has gained the interest of numerous research groups in the past. However, most of the aforementioned works have focused on the entrapment and rising of bubbles in yield stress fluids; there is barely any information about the mass transfer of bubbles in yield stress fluids, particularly, on a microscale point of view. In this work, the motion of a single bubble and its mass-transfer process to the surrounding yield stress fluids is simulated using the VOF method for tracking the bubble interface.

RESULTS AND DISCUSSION

Change in the Concentration Field around a Rising Bubble. Fundamentally, the mass-transfer process of a single bubble rising in yield stress fluid is simulated, which can be observed from Figure 1 that with the rise of a single bubble, the "tail" formed through the concentration field behind the bubble becomes longer. Nevertheless, it will not modify when the wake grows to a certain length. On the one hand, when the bubble rises, the friction between the bubble interface and the liquid phase causes the gas phase inside the bubble to change from being static to slowly form a vortex along the wall from the outside to the inside, and the surrounding air flow meets in the middle of the bottom of the bubble, which thereafter goes up along the center of the bubble, inside the bubble, and top of the bubble, ultimately forming a loop. Accordingly, the air flow at the bottom of the bubble gathers little by little in the middle. In addition, during the bubble rise, the liquid phase near the interface flows from the head of the bubble, along the bubble interface to the tail of the bubble, and eventually gathers in the middle, compressing the concentration field into this shape. However, the bubble stabilizes by degrees as it rises, and the length of the tail does not change.

Influence of the Bubble Diameter and Fluid Properties on Mass Transfer. Figure 2 shows the effects of bubble diameters on the CO₂ concentration profiles inside the liquid phase near the bubble equator. It can be seen from Figure 2 that the thickness of the concentration film increased with the increase of the bubble radius. The bubble has a larger velocity and a stronger capability to surmount the yield stress when the radius of the bubble becomes larger. In addition, large bubbles possess greater velocities, so the gas–liquid phase near the interface has a greater effect on the velocity and the gas–liquid phase has a strong effect on the concentration near the interface and the thickness of the concentrated boundary layer. With the increase of the bubble radius, the velocity of the gas–liquid phase increases, which extremely boosts the transfer of CO₂ from the gas phase to the liquid phase, resulting in a significant increase of the liquid phase near the boundary and the thickness of the concentration boundary layer. Komori investigated the relationship between the mass transfer coefficient of the liquid phase and the shear rate at the gas–liquid interface, and caught sight of that the mass transfer coefficient increased with dispatch with the shear rate increases, and the inquiry consequences were similar to those in this paper.

Figure 3 shows the concentration distribution of a single rising bubble near the equator in a yield stress fluid. Moreover,
The effect of yield stress on the CO₂ mass fraction field is shown in Figure 4. The results illustrate that the dissolved CO₂ is distributed behind the bubble and forms a straight line in the liquid phase, but the mass fraction field line becomes thinner and longer as the yield stress decreases because of the inhibition of yield stress in the gas–liquid phase mass transfer. Because of the high yield stress, the CO₂ concentration in the bubble wake zone does not easily diffuse into the surrounding liquid phase. Therefore, the concentration line in the wake zone is thinner and longer.

The CO₂ concentration field in yield stress fluids with different consistency coefficients (K) is shown in Figure 5. The results illustrate that the dissolved CO₂ line behind the bubble decreases with increasing consistency coefficients (K) because of the high viscosity of the large consistency coefficients, which is harmful to the gas–liquid phase mass-transfer process.

As depicted in Figure 6, the variation of concentration distribution at the equator of a single rising bubble in yield stress fluids with different consistency coefficients (K) is compared, which clearly showed that at the same distance from the bubble interface, the concentration of CO₂ showed an apparent decrease when the consistency coefficient K increases. In the Herschel–Bulkley (H–B) model, the consistency coefficient of fluids prevalently mirrors the characteristics of viscosity in yield stress fluids; the larger the consistency coefficient is, the greater the viscosity of the solution and the smaller the diffusion coefficient of CO₂ in the liquid phase, which is not conducive to mass transfer between the gas and the liquid.

The flow index (n) in the H–B model quantitatively portrays the degree to which a non-Newtonian fluid deviates from a Newtonian fluid. When the flow index n equals 1, the viscosity μ in the power law model is identical to the consistency coefficient K, and this type of fluid is referred to as the Newtonian fluid. When the flow index n is less than 1, this kind of fluid exhibits a shear thinning behavior. When n surpasses 1, the fluid mirrors the property of shear thickening. The yield stress fluid used in this paper exceeds the critical yield stress, which is indicative of shear thinning characteristic and can be considered as a shear thinning fluid. Therefore, this paper principally makes enquiries on the mass-transfer process in bubble rising when the flow index n is less than 1. The effects of flow index (n) on the CO₂ concentration field are shown in Figure 7, which indicates that the dissolved CO₂ line behind the bubble increases with the increase of the flow index (n). Figure 8 shows the change in the liquid phase concentration distribution at the bubble equator in yield stress fluids with the flow index n equal to 0.5, 0.7, and 0.9. Legible as the Figure 8 seems, the concentration of CO₂ at an equal distance from the bubble interface decreases with the increase
of the flow index. However, with the decrease of the flow index, the local viscosity around the bubble decreases at the same shear rate, such that the viscous resistance of mass transfer between the gas and liquid excessively decreases as well.

**Bubble Drag Coefficient and Mass-Transfer Correlation in Yield Stress Fluids.** The forces acting on the bubble are principally the buoyancy and drag force. Based on the second law of Newton

\[
(\rho_l - \rho_g) g \frac{\pi d_b^3}{6} = C_D \frac{1}{2} \rho_l U_b^2 \frac{\pi d_b^2}{4}
\]

where \( \rho_l \) and \( \rho_g \) are the density of the liquid and the gas, respectively. By neglecting the gas density, the bubble drag coefficient \( C_D \) can be written as follows

\[
C_D = \frac{4 \rho_g}{3 U_b^2}
\]

and the Reynolds number can be defined as

\[
Re = \frac{\rho U_b d_b}{\mu}
\]

The viscosity of the liquid around the bubble can be calculated using eq 30, where \( \dot{\gamma} \) is calculated as \( \dot{\gamma} = U_b/d_e \). Thus, the Reynolds number is defined as follows

\[
Re = \frac{\rho U_b^{2-n} d_e^n}{K}
\]

For a single bubble with a shear-free interface, the drag coefficient equation can be calculated by the Hadamard–Ribczynky (H–R) model

\[
C_D = \frac{16}{Re}
\]

**Figure 9** illustrates the comparison of the computed drag coefficients and correlations taken from eq 5. It distinctly clarifies that the simulated results do not agree with the H–R model because of the yield stress. Thus, the yield stress behavior of non-Newtonian fluids should be fully considered apart from \( Re \) to establish a more precise correlation of the drag coefficient. Blackery\(^{27}\) used the finite element method to evaluate the total drag on a sphere moving slowly (creeping regime) in yield stress fluids and forward predictions in terms of the correction factor \( X (= C_D Re/24) \), which now becomes a function of the Bingham number as

\[
X = 1 + aBi^b
\]

where the Bingham number is defined as

\[
Bi = \frac{\tau_y d_e^{n-1}}{KU_b^{n}}
\]

Based on the above consideration, a comprehensive correlation was obtained by fitting the 81 simulated data

\[
C_D = \frac{16}{Re} (1 + 1.35Bi^{0.86})
\]

The average relative deviation between the forecasted and simulated total drag coefficient is about 20%, which is acceptable for the complex rheological behavior of yield stress fluids. The validity of eq 8 for single bubbles in yield stress fluids is also shown by plotting \( C_D/(1 + 1.35Bi^{0.86}) \) in **Figure 10**. This figure covers the ranges 0.029 < \( Re \) < 8.54 and 0.35 < \( Bi \) < 6.89. In these ranges, the present correlation approximates are in good agreement with simulated data.

**Figure 9.** Comparison of the computed drag coefficients and the H–R model (\( C_D = 16/Re \)).

**Figure 10.** Correlation of transient drag coefficients against Reynolds numbers.

The liquid side mass-transfer coefficients are related to fluid properties, characteristic sizes, and the relative velocity of the bubble and the liquid phase. Levich\(^{28}\) obtained a correlation for mass transfer from a single bubble as follows

\[
Sh = 0.65Pe^{1/2}
\]

where the Sherwood number (\( Sh \)) and the Peclet number (\( Pe \)) are defined, respectively, as follows

\[
Sh = \frac{k_L d_e}{D_A}
\]

\[
Pe = \frac{d_e U_b}{D_A}
\]

where \( k_L \) and \( D_A \) could be obtained from eqs 28 and 29, respectively.

**Figure 11** shows the comparison of the computed Sherwood numbers (\( Sh_L \)) from eq 9 and the simulated Sherwood numbers (\( Sh_s \)). The simulated Sherwood numbers (\( Sh_s \)) and the computed Sherwood numbers (\( Sh_L \)) from eq 9 are not consistent. Some reports concluded that the yield stress showed a decrease of mass-transfer rate.\(^{29,30}\) Therefore, the
The mass-transfer behavior of bubble rising in yield stress fluids is investigated numerically by using VOF and UDF (user-defined function) methods. The CO₂ concentration profiles inside the liquid phase near the bubble equator are used to investigate different bubble diameters, yield stresses, consistency coefficients, and flow indices. In addition, the thickness of the concentration film near the bubble equator can indicate the rate of mass transfer. The conclusions are corroborated by the fact that the rate of mass transfer decreased with an increase of yield stress, consistency index, and flow index of the liquid phase and with a decrease in bubble diameter. The simulated drag coefficients and Sherwood numbers are compared with the values calculated by the classical H—R model (\(C_D = 16/Re\)) and the Levich model (\(Sh = 0.65Pe^{1/2}\)), respectively, and the results point out that the simulated drag coefficients are higher than those calculated using the H—R model line and the simulated Sherwood numbers are smaller than the calculated ones with the Levich model because of the yield stress. In order to correlate the deviation which is caused by yield stress, a correction factor \(X\) for \(C_D\) correlation and another correction factor \(f_c\) for Sherwood numbers are introduced, which can be expressed as a product of Bingham number (\(Bi\)). The present correlations are in good agreement with simulated statistics.

### FORMULATION

**Governing Equations.** CFD software FLUENT is utilized to simulate the motion and the mass-transfer process of a single bubble rising in yield stress fluids, and the bubble interface tracking method is based on the VOF approach. These simulations are carried out with the following basic assumptions: (1) the gas phase and liquid phase are incompressible, (2) the flow process is isothermal, and (3) the flow process is laminar. The bubble rising in yield stress fluids could be characterized by the following continuity and Navier—Stokes equations.

The continuity equations can be written as

\[
\nabla \cdot \mathbf{u} = 0
\]

where \(\mathbf{u}\) is the fluid velocity vector.

The Navier—Stokes equations considering the surface tension can be expressed as

\[
\rho(F) \left( \frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla p + \nabla \cdot \mathbf{\tau} + \mathbf{F}_s + \rho(F) g
\]

where \(p\) is the pressure, \(\mathbf{F}_s\) is the surface tension, \(\rho(F)\) is the density, \(\mathbf{\tau}\) is the stress tension, and \(g\) is the gravitational acceleration. Moreover, \(\mu(F)\) and \(\rho(F)\) can be defined as

\[
\mu(F) = \mu_1(F) + \mu_0(1 - F)
\]

\[
\rho(F) = \rho_1(F) + \rho_0(1 - F)
\]

where \(\mu(F)\) is the coefficient of kinematic viscosity and subscripts \(g\) and \(l\) denote the gas phase and the liquid phase, respectively. \(F\) is the volume fraction, which can be written as

\[
F = \begin{cases} 
0 & \text{gas region} \\
0 < F < 1 & \text{interface} \\
1 & \text{liquid region}
\end{cases}
\]

The volume fraction \((F)\) should be satisfied by the following equation

\[
\frac{\partial F}{\partial t} + \nabla \cdot (\mathbf{u} F) = 0
\]

For yield stress fluids, the stress tension \(\tau\) could be calculated using the H—B model as follows
\[ \tau = \tau_0 + K(\dot{\gamma})^n \]  
(20)

where \( \dot{\gamma} \) denotes the shear rate, which should be computed at each nodal point of the computational domain. Thus, the shear rate \( \dot{\gamma} \) changes with the velocity vector instantaneously and can be calculated by the following formula \(^{31}\)

\[ \dot{\gamma} = \sqrt{2\left(\nabla \cdot \mathbf{D}\right)^2} = \sqrt{2\tau(rD^2)} \]

\[ = \sqrt{2\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} + \frac{1}{2} \frac{\partial u_i}{\partial y} + \frac{\partial u_j}{\partial y}\right)^2} \]

\( D \rightarrow \) is the strain rate tensor.

**Momentum Source Term Due to Surface Tension.** The term \( F_s \) is introduced in the momentum following the continuum surface force model of Akita and Yoshida \(^{35}\) as

\[ F_s = \sum_{i<j} \sigma_{ij} \rho \kappa_i \nabla F_i + \rho \kappa \nabla F_i \]

\[ \frac{\partial}{\partial t}(\rho \vec{v}) + \nabla \cdot (\rho \vec{v} \vec{v}) = \nabla \cdot (\lambda \nabla \vec{v}) + \rho \vec{g} + \vec{f} \]

\[ = \left[ \frac{\partial \kappa_i}{\partial x_j} \right] \nabla \cdot (\rho \vec{v} \vec{v}) + \rho \vec{g} + \vec{f} \]

where \( \sigma \) is the surface tension and \( \kappa \) is the local surface curvature, which could be calculated as follows

\[ \kappa = -\left(\nabla \cdot \vec{n}\right) = \frac{1}{|\vec{r}|^2} \left[ |\vec{r}| \nabla |\vec{r}| \right] - (\nabla \cdot \vec{r}) \]

\( \vec{n} \) is the unit normal vector of the interface.

**Species Conservation Equation.** The equation describing the transport of species is given by

\[ \frac{D(\phi_i \rho c)}{Dt} = D_i \nabla^2 (\phi_i c) + S_m \]

where \( S_m \) is the source term, \( \phi_i \) is the percentage composition of the liquid phase, \( c \) is the molar concentration, and \( D_i \) is the molecular diffusion coefficient.

The mass-transfer source term of the bubble to the liquid phase is expressed as follows

\[ S_m = \alpha_i k_i (c_i - c_{\infty}) \]

where \( \alpha_i \) is the specific surface area, \( c_i \) is the concentration in the liquid phase near the interface, \( c_{\infty} \) is the concentration in the liquid phase, and \( k_i \) is the liquid-phase mass-transfer coefficient, which can be calculated using Higbie’s penetration model

\[ k_i = 2 \frac{D_i}{\pi \mu c} \]

\( t_c \) is the exposure time, which can be calculated by the slip velocity of a bubble. For a single bubble, the exposure time is estimated using the ratio of the bubble velocity and the diameter. Thus, the \( k_i \) for a single bubble is defined in the following equation as

\[ k_i = 2 \frac{DU_i}{\pi \Delta d} \]

\( \Delta d = \frac{2J}{\pi \Delta d^3} \)

where \( \Delta d = \frac{2J}{\pi \Delta d^3} \). The concentration in the bubble interface is given by

\[ c = c_i \]

The operational operating pressure is set as 1 atm in the coordinates \( x = 20 \text{ mm} \) and \( y = 10 \text{ mm} \). The pressure outlet boundary condition is employed at the top of the computational domain. The pressure is fixed as follows

\[ p = 0 \]

The time step \( \Delta t \) is \( 1 \times 10^{-3} \). In this paper, the triangle mesh is used and the size is set to 0.5 mm, which could ensure the accuracy of bubble motion simulation according to our previous examination (Table 1) \(^{35}\).

| Table 1. Computational Conditions |
|----------------------------------|----------------|----------------|
| property                         | Value          |
| equivalent diameter of the bubble, \( d_e \) mm | 6              | 9              | 12            |
| yield stress, \( \sigma \), Pa    | 3              | 5              | 7             |
| consistency coefficient, \( K \), Pa-s\(^n\) | 2              | 4              | 6             |
| flow index, \( n \)               | 0.5            | 0.7            | 0.9           |

**Experiment and Model Validation.** Validation of the simulation is carried out through a comparison of the experimental and simulated results, which are presented in Table 2 and composed of 27 cases. The comparison results in Table 2 demonstrate a good agreement between the computational and experimental results. These simulations
suggest that our method is effective for investigating the bubble motion in yield stress fluids.

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Notes
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