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Review

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The interphase mass transfer in liquid–liquid systems with Marangoni effect

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Abstract. In this paper, we review the related studies on the interphase mass transfer process accompanied with Marangoni effect in liquid-liquid systems. The Marangoni effect is triggered by the local variation of interfacial tension and influenced by many factors, such as the physicochemical properties of the system, the solute concentration and the bulk flow. The onset criterion of the Marangoni effect has been discussed extensively via theoretical analysis and experimental verification, but a unified and universal criterion was still not developed due to the complex system geometry and boundary conditions. When the Marangoni convection occurred, the bulk flow adjacent to the interface was spontaneously disturbed, normally leading to an enhanced mass transfer coefficient. Besides, the surface active agent has been found to affect the solute transport across the interface, by either promoting or inhibiting the Marangoni convection according to the nature of additives.

1 Introduction

The Marangoni effect is usually encountered in mass/heat transfer in multiphase reactors, extraction columns and distillation towers, and it manifests itself in diversified forms of interfacial behaviors including surface rippling, localized eruption, interfacial turbulence and fluid film oscillation [1]. The additional effect on mass/heat transfer efficiency and product quality has attracted chemical engineers' attention for more than 50 years. The research of Marangoni effect is expected to contribute to further exploration on the mechanisms of interphase mass transfer.

The inhomogeneity of temperature and/or solute concentration can generate local surface tension gradient which tends to trigger Marangoni effect. The thermal Marangoni effect, also termed as thermocapillary instability, has been studied extensively, and several excellent reviews [2–5] may be referred. However, this work is focused on the current researches of Marangoni effect induced by interphase mass transfer in liquid-liquid extraction systems. The solutal Marangoni effect is more complex than the thermal counterpart in three main aspects. (a) The interphase mass

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transfer theory is still premature. The classical models, including the double-film theory, the penetration theory, the surface renewal theory, and the later developed ones, such as the vortex cell model [6] and the vortex diffusion model [7], cannot yet fully reveal the essence of interphase mass transfer. (b) In contrast to the thermal impetus that can be artificially controlled as a constant, the mass transfer is usually a transient process, and the driving force dissipates along the time. (c) In a contaminated system, surfactants may be accumulated on and transported along the interface, which requires extra sophisticated modeling.

A set of nonlinear partial differential equations, consisting of the Navier-Stokes equation, continuity and mass conservation equations, can be used to describe the Marangoni effect. When the two liquid phases contact, the force balance over the interface must be satisfied. This momentum transfer is thus coupled with the mass transfer by the interface conditions, in equilibrium or non-equilibrium. According to the basic principles of hydromechanics, the boundary condition of the two-phase momentum balance is written in the following general form [8]:

$$\left[p^{(2)} - p^{(1)} + \sigma \left(\frac{1}{r_1} + \frac{1}{r_2}\right)\right] n_i$$

$$= \left[\mu^{(2)} \left(\frac{\partial v_i^{(2)}}{\partial x_k} + \frac{\partial v_k^{(2)}}{\partial x_i}\right) - \mu^{(1)} \left(\frac{\partial v_i^{(1)}}{\partial x_k} + \frac{\partial v_k^{(1)}}{\partial x_i}\right)\right] n_k - \frac{\partial \sigma}{\partial x_i}$$
(1)

where p, μ and v_i (i, k = 1, 2, 3) are respectively pressures, dynamic viscosities and velocity components in two phases marked as superscripts (1) and (2), r_1 and r_2 are the principal radii of curvature of the surface, n_i is the components of the outward unit vector normal to the surface and directed into the interior of phase 1, σ is the surface tension, which is a variable dependent on solute concentration as well as surfactant distribution at the interface. For describing systems with surfactants, on the right-hand side of Eq. (1) a term should be added:

$$-\mu_{\rm S}\delta_{ik}\frac{\partial^2 v_k^{\rm (s)}}{\partial x_i \partial x_k} \tag{2}$$

where δ_{ik} is the Kronecker symbol, μ_s is the surface viscosity, and superscript (s) denotes the surface. This term accounts for energy dissipation caused by the irreversible processes (such as the accompanying deformation of free interface) occurring on the interface. The surface viscosity, which cannot be measured directly in experiment, is usually calculated by the physical model and verified by the experimental data.

The above description indicates that the Marangoni effect is a typical nonlinear and complicated phenomenon in engineering. It can be linearized by the small perturbation analysis to study and forecast approximately the marginal Marangoni phenomena. The weakly nonlinear analysis, breaking through the limit of linear stability analysis, can provide the prediction of flow pattern and the evolution of Marangoni effect. These theoretical methods with experimental validations have made definite contributions for understanding the Marangoni effect [9-12]. With the development of the computer technology and numerical methods, the direct numerical simulation has become a valuable and reliable tool to perform the engineering researches, as it can be applied in cases with much more complex boundary conditions and give an overall view of flow fields coupled with mass transfer processes. A few numerical methods have been proposed so far to solve the full Navier-Stokes equations with a deformable interface, such as the lattice Boltzmann method [13], the phase-field method [14], the volume of fluid method [15], and the level set method [16]. The modeling of surface evolution is perfectly developed in these numerical algorithms. The virtual interface, approximately one or two grid points, is usually utilized in the numerical computation to smooth the discontinuity of parameters between two fluids, leading to the loss of the flow information on small scale (smaller than the virtual interfacial thickness) and the artificial diffusion in the numerical simulation. Meanwhile, the spurious parasite currents are similar in appearance with the interfacial turbulence. Hence, the gap is often wide between the predicted results and the experimental data of the transient evolution of the Marangoni effect. Whether the results are consistent with the natural laws should be demonstrated by reliable experiments.

The direct evidences can be caught by the optical photographic devices like the Schlieren technique [17, 18], the particle image velocimetry (PIV) [19, 20], and the realtime dynamic holographic interferometry [21,22], which have been developed rapidly in recent decades. They provide convenient conditions for investigating the interfacial instability occurring during interphase mass transfer processes. As the optical observation requires a clear phase interface, the experimental technology is restricted in a static or strictly laminar liquid-liquid interface, laminar falling liquid film or a quiescent drop in an immiscible continuous phase. In addition to direct observation, some parameters can also be used to identify whether the Marangoni effect occurs or not. For instance, the drop velocity will decrease because the Marangoni convection destroys both the internal and external flow along the interface and increases the drag coefficient. Wegener et al. [23] found a two-stage acceleration behavior and the temporary reduction of drop rise velocity. These phenomena offered a wealth of useful signals of the interfacial convection to this area. The mysterious veil of the Marangoni effect is gradually unraveled by integrated application and incorporation of the above-mentioned research methodology.

2 Onset and development of the Marangoni effect

The infinitesimal fluctuations of solute concentration at the liquid-liquid interface create a local concentration gradient, and it leads to a surface tension gradient which tends to contract or stretch the interface. Therefore, the liquids at both sides are subjected to some tangential force, which induces surface deformation and bulk phase flows. If the conditions allow the infinitesimal patterns of interfacial flow to develop into finite ones, they will become visible, resulting in the enhanced mass transfer process between two phases. Hence, in addition to the properties of the solvent (kinematic viscosity ratio, solute diffusion coefficient ratio, and interfacial tension sensitivity) of a specific system, many other factors (mass transfer direction, geometry constraints of interface) may become a fatal parameter to provoke the Marangoni convection.

Sternling and Scriven [24] adopted the linear stability analysis to deduce the stability criterion of the interfacial instability on a non-deformable planar interface with a solute transferred between two immiscible liquids. The system in which the interfacial tension was sensitive to the solute concentration was prone to the Marangoni effect, when the solute transfer occurred from the phase with a higher viscosity and a lower diffusivity. The stability analysis of solutal Marangoni convection has been extended by Hennenberg et al. [25–27] with consideration of non-linear concentration profiles, spherical interface, effect of adsorption and desorption etc. Chu and Velarde [28] obtained instability criteria for three different types of waves, not only the longitudinal but also transversal modes of oscillation. Nakache et al. [29] proposed a semi-empirical criterion for surface active agents transfer accompanied with the solutal Marangoni effect in ternary liquid–liquid extraction. Unfortunately, a poor agreement was encountered when the above-mentioned theoretical predictions were compared with the experimental investigations [30] of the interfacial instability on 35 different liquid-liquid interfaces. A new Marangoni number was defined with the surfactant molecular mass for characterizing different kinds of surfactants, and accurate predictions of the systems in their experiments were reported. However, whether the critical Marangoni number is a universal criterion or not still needs more stringent verifications. Based on the nonlinear dynamics, Kovalchuk and Vollhardt [31] explained the oscillation mechanism in the mass transfer process of a nonionic surfactant through liquid/liquid interface with nonlinear dynamics, and presented an oscillation criterion which the linear analysis failed to predict. The direct numerical simulations [13, 32, 33] of the liquid-liquid extraction system is not limited by the basic assumptions in linear and nonlinear methods, and contrarily, it becomes a useful tool to estimate the real Marangoni convection with good performance. In addition, a brand new phenomenon was discovered by the Monte-Carlo simulations performed by Schott and Pfennig [34] in the lattice system. That is, the nano droplets formed in the close vicinity of the interface, which has not been caught in the experimental investigations to date. This could be a good explanation of the eruption and turbulent movement at the liquid-liquid interface.

The evolution of the flow patterns was fascinatingly concerned in exploration of the Marangoni effect. Sternling and Scriven [24] attributed the interfacial convection to the non-uniform distribution of solute on the interface and predicted the roller convective structure. Schwarzenberger et al. [35] reviewed the stationary solutal Marangoni instability in two planar liquid systems and described in details the instability evolution in forms of roll cells, chaotic relaxation oscillations and synchronized relaxation oscillations. Based on the level set method, Wang et al. [36] simulated numerically the deformable drop driven by buoyancy in an infinite continuous phase. The roll cells appeared initially, very small and dense-packed adjacent to the interface and then propagated along the interface. Finally the roll cells decayed and the internal circulation re-emerged. By analyzing the drop behaviors, Wegener et al. [37] proposed an evolution model of the Marangoni effect on drop scale to explain the drop deviation from its original vertical pathway. This model (as showed in Fig. 1) revealed that a transition period existed between the fully developed Marangoni convection and the internal circulation. In the transition period, the internal circulation reshaped, however, with an arbitrary choice of the location.

The solute-induced Marangoni effect will take some time to develop instable convection adjacent to the interfacial boundary. The numerical simulation by Mao and Chen [38] demonstrated that the Marangoni effect occurred only when the concentration boundary layer became thick enough. Bushueva et al. [39] observed the Marangoni effect on the surface of the chlorobenzene droplet and revealed the mass transfer rate of the surfactant confined the onset of the Marangoni effect. In a contaminated system, the surface tension gradient occurs until the surfactant is adsorbed onto the interface and is transported along the interface. Javadi et al. [40] also investigated a significant delay in the onset of the Marangoni effect after the concentrated surfactant solution reached the drop surface, and demonstrated this phenomenon with the probable mechanism within the following aspects: the interfacial layer barrier, a kinetics-controlled adsorption mechanism, a critical Marangoni number and the renewal mode of deformable drop interface.

The modeling of the solutal Marangoni effect is already in a mature state. Hence, the numerical technology can take the nonlinear problem of the Marangoni effect into full consideration and give a relatively accurate prediction of the Marangoni convection. However, it seems that the simulation processes are more likely to predict an earlier onset of the Marangoni effect [36,40]. The existence of the spurious parasite currents and the artificial diffusions, imported by the numerical discretization of the

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Fig. 1. Main stages of schematic Marangoni convection flow pattern in a rising droplet against corresponding concentration field from numerical simulation of an acetone laden 2 mm toluene droplet rising in water: (a) early stage, large concentration gradients, strong Marangoni convection, enhanced radial mixing and fast mass transfer, (b) middle stage, decreasing strength of Marangoni convection, asymmetric onset of internal circulation, (c) later stage, no Marangoni convection, symmetric toroidal internal flow field [37].

surface tension model, promotes and strengthens the interfacial turbulence. Moreover, the numerical model still remains unclear on the interface mechanisms, including the description of the interfacial layer thickness and the renewing mechanism of the deformable interface. So, it leaves a difficult job to investigate the Marangoni effect in different experimental systems. The experimental results can be used to verify the current numerical models. For example, the experimental data for the evolution of the Marangoni effect on an unsteady drop is still absent as yet.

3 The Marangoni effect enhanced mass transfer coefficient

The occurrence of Marangoni effect can accelerate the renewal of interface, strengthen the bulk phase mixing, and dramatically promote the mass transfer efficiency. The mass transfer coefficient in the system with an unstable interface is critical for the further research of interphase mass transfer mechanism, since it has been reported significantly higher than that predicted by the two-film and penetration theories. Moreover, the enhanced mass transfer coefficients by the Marangoni effect is much more concerned by the chemical engineers for it will be an important parameter to the design and scale-up of industrial extraction devices.

Based on the three classical models of interphase mass transfer, a series of methods were reported to predict the mass transfer coefficients from a single rigid drop to an infinite continuous phase. These theoretical and empirical equations (as listed in Table 1) have different hypotheses of the internal flow patterns and ignore the mass transfer resistance in the continuous phase. Assumed that stagnancy inside the drop and mass transport only by pure diffusion, Newman [41] derived the internal stagnant drop model. For a long contact time, the model could be reduced to a constant mass transfer coefficient: $k_{\rm d} \cong 6.6$. Kronig and Brink [42] considered a spherical drop with

Authors	Models
Newman [41]	$k_{ m d} = -rac{d_{ m e}}{6t} \ln\left[rac{6}{\pi^2}\sum_{j=1}^\infty rac{1}{j^2} \exp\left(rac{-4j^2\pi^2 tD_{ m d}}{d_{ m e}^2} ight) ight]$
Kronig and Brink [42]	$k_{ m d} = -rac{d_{ m e}}{6t} \ln \left[\sum_{j=1}^{\infty} rac{3}{8} B_j^2 \exp \left(rac{-64 \lambda_j t D_{ m d}}{d_{ m e}^2} ight) ight]$
Handlos and Baron [43]	$k_{ m d} = -rac{d_{ m e}}{6t} \ln \left[2 \sum_{j=1}^{\infty} C_j \exp \left(rac{-16\lambda_j t U_{ m T}}{2048 d_{ m e} \left(1 + \mu_{ m d}/\mu_{ m c} ight)} ight) ight]$

Table 1. Models to predict mass transfer for a rigid drop.

internal circulation following the Hadamard streamlines with the Reynolds number smaller than 0.25. Compared with the Newman equation, the mass transfer coefficient was enhanced more than two times due to the internal circulation. To describe the internal mixing, Handlos and Baron [43] added a turbulence-like disturbance to the internal circulation.

These models listed in Table 1 take no account of the Marangoni effect, but can be used to determine the influence of the Marangoni convection on the mass transfer coefficients. Sawistowski and Goltz [44] observed that when the Marangoni effect occurred the mass transfer coefficient was higher than that predicted by the penetration theory and the mass transfer rate was a function of both the interfacial tension and the solute concentration in the donor phase. The extraction fractions measured by Wang et al. [45] for the solute transferred from a hanging drop (1-hexanol) to the aqueous phase was always higher than those predicted by the stagnant model and broadly agreed well with those predicted by the Kronig-Brink model. However, for the cases with higher solute concentrations, the Marangoni developed progressively, and the extraction fraction was much higher than the Kronig-Brink model prediction. If the parameter related to the Marangoni effect is incorporated, these models can be improved to predict the mass transfer coefficients with Marangoni effect. We gener and Paschedag [46] performed a series experiments to investigate the Marangoni effect on mass transfer from a single organic drop to the aqueous phase. Their results show that the Handlos-Baron model could predict the Marangoni dominated mass transfer process only in the right trend. The Handlos-Baron model was improved by introducing the initial solute concentration difference between two phases to characterize the intensity of the Marangoni effect and then it agreed very well with the experimental data. Zheng et al. [47] adopted an enhancement factor, reflecting the effect of interfacial instability, drop rise velocity and initial solute concentration, to modify the improved Newman model [48]. Moreover, they predicted the mass transfer coefficients of the *n*-propyl acetate/acetic acid/water system with relative error of 20%.

Grahn [33] offered a 2D numerical simulation of two horizontal liquid layers and the concentration distribution normal to the interface. It was indicated that the mass transfer in the interfacial vicinity was dominated by the molecular diffusion. Compared with the Rayleigh-Bénard effect induced by the density gradient, the Marangoni effect can accelerate the convection only in the area close to the interface and has limited impact in the area deep into the bulk flow. Due to the small confined space inside the drops, it is readily to deduce that the Marangoni convection will have much more significant influence on the internal flow and the drop behavior. The experimental investigation of the toluene/acetone/water system by Wegener et al. [49] showed that the mass transfer rate decreased and later increased along with the initial solute concentration. At a lower initial solute concentration, the weak interfacial convection disturbed the internal circulation and reduced the drop velocity relative to the continuous phase, depressing the mass transfer rate. As the initial concentration increased, the Marangoni effect became strong enough for compensating the absence of the internal circulation. The drop motion slowed down and the mass transfer rate was accelerated. However, both the experimental data and the numerical results [36] of single deformable drop extraction in the MIBK (methyl isobutyl ketone)/acetic/water system indicate that the mass transfer coefficients were larger with higher initial solute concentrations. Mao and Chen [38] analyzed four virtual cases with different molecular diffusion ratios and kinematic viscosities, and found that the mass transfer coefficients were not always enhanced but possibly decreased a little with occurrence of the Marangoni effect [38]. Their numerical results [38] demonstrate that the vortices developed along the outer surface of the drop destroyed the laminar flow around the drop, and further contributed to the mass transfer resistance in the continuous phase. All the above facts suggest that the Marangoni effect will have a significant influence on the mass transfer process in a complex way, which is decided by a comprehensive coupling of interfacial instability, internal circulation, drop behavior and the intrinsic properties of the system.

4 Marangoni effect in a contaminated system

In most industrial extraction equipments, unavoidable trace quantities of surface active contaminants may have a profound effect on interphase mass transfer. An insoluble surfactant may accumulate on the phase-contacting area and transport along the interface [50]. The effect of an insoluble surfactant was investigated by Lee and Pozrikidis [51] via introducing the immersed-interface method and the diffuse-interface approximation. Xu et al. [52] established an Euler method for the transport and adsorption of the insoluble surfactant on the interface. As for a soluble surfactant, the absorption from and desorption to the bulk phase should be incorporated as the rate of change of the interfacial excess in the convection-diffusion equation of the surfactant. Based on the diffuse-interface method, Teigen et al. [53] proposed a numerical algorithm for a soluble surfactant mass transfer process coupled with adsorption on the interface. Li et al. [54, 55] studied the influence of surfactants on the buoyancy-driven drop motion and interphase mass transfer in the orthogonal boundary-fitted coordinate system under the condition of constant interfacial tension.

Extensive theoretical and experimental researches of the liquid-liquid extraction attributed the effect of surfactants on the mass transfer process for the fluid mechanics and the molecule effect. From the fluid mechanics perspective, the majority of researches [56, 57] show that the surfactant could suppress the interfacial convection and the internal circulation, being a hindrance to the liquid-liquid extraction. A reasonable explanation for this is that the surfactant adsorbed on the interface makes the drop surface more rigid. Arendt and Eggers [58] discovered that in the toluene/acetone/water system the surfactant (Triton X-100) weakened the interfacial instability because of the formation of the rigid surfactant monolayer. The recent investigations indicated that the surfactant could also encourage the interfacial instability and enhance the mass transfer coefficients. Agble and Mendes-Tatsis [30] observed the interfacial phenomena in the water-organic reagent system with six surfactants and found that the ionic surface active agent tended to stimulate the Marangoni effect, but the nonionic surfactant had little or negative effect on the interfacial convection. Wang et al. [45] found that both ionic and nonionic surfactants dampened solute interfacial convection at low surfactant concentrations (as showed in Fig. 2). However, SDS (sodium dodecyl sulphate) introduced intensified interfacial



Fig. 2. Effect of surfactant on interfacial instability of a 1-hexanol drop hanging in aqueous phase with interphase mass transfer [45].

instability while no interfacial instability was observed for Triton X-100. The presence of SDS in high concentration induced the intensified interfacial instability accompanied with drop oscillation, and the extraction fraction enhancement factor was up to 3.37-6.3. Meanwhile, when the concentration of Triton X-100 was increased to $0.5 \,\mathrm{g/L}$, the interfacial instability was triggered like a smoke emission from the interface without any drop oscillation. The extraction fraction was still lower than that of the stagnant model. The authors suggested that the decrease of mass transfer rate resulted from the interfacial resistance generated by the surfactant monolayer adhered on the interface. The solute molecules transferring between two phases had to overcome the interaction with surfactant molecules. When the instability was too weak to counteract the increased interfacial barrier, the mass transfer rate would be decreased. Hutchinson [59] determined the mass transfer rates for different solutes in the water/benzene system with dilute sodium hexadecylsulphate, and found that the hydrophilic groups added the interfacial resistance for the solute transfer. When the acetic acid transferred from a CCl₄ drop to the continuous water phase, the nonionic surfactant Triton X-100 and the anionic surfactant SDS decreased the mass transfer coefficient to 7% of that in the pure system. The cationic surfactant DTMAC reduced the mass transfer coefficient to 30% of that in a pure system [60], where the inhibition effect on the mass transfer was equivalent for different types of surfactants. The authors deduced that the interfacial resistance generated by adsorption of surfactant played a major role in the mass transfer process.

5 Conclusion

The progress on researches of the solute-induced Marangoni effect occurring in the liquid-liquid extraction is reviewed in this work. The numerical simulations and experimental equipments are being developed rapidly in recent years even with a preliminary attempt in the three-dimensional framework. However, the onset criterion of the Marangoni effect still needs further modification, since it is a complex nonlinear problem and develops transiently in both space and time. The numerical simulation has not been verified fully for its reliability and cannot provide a perfect prediction of the interfacial instability as an independent tool. The modeling of the two-phase flow deserves further improvement on the accurate description of the deformable interphase surface. On the other hand, the experimental measurement of the Marangoni effect needs a more sophisticated design to obtain the detailed flow information on the interface. Besides, three-dimensional determination (with the help of digital holographic particle image velocimetry or the optical coherence tomography etc.) of the two-phase flow would be very beneficial to investigate the onset of the Marangoni convection and the local flow field near the interfacial boundary. Furthermore, the three-dimensional experimental data are also needed for the validation of numerical simulations.

The effect of the Marangoni convection on the mass transfer process was a result of comprehensive consequence of the initial solute concentration, the drop behavior, the surfactants and so forth. A large volume of data is necessary to screen out the mechanism of the solute-induced Marangoni effect. The current studies are frequently focused on the average mass transfer coefficients. However, the local concentration variation would illustrate essentially the mass transfer mechanisms. Therefore, a real time concentration measurement like the holographic interferometer or the planar laser-induced fluorescence should be applied to investigate the solutal Marangoni effect. Moreover, the numerical simulation is also to be improved to eliminate the artificial diffusion and get the exact concentration field, especially in the interfacial area.

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Symbols

B	eigenvalue in Kronig-Brink model and Handlos-Baron model
d_e	volume-equivalent diameter of drop, m
D	diffusion coefficient, $m^2 s^{-1}$
k	mass transfer coefficient, m s^{-1}
n	unit vector component
p	pressure, Pa
r	principal radii of curvature, m
t	time, s
v	velocity, m s^{-1}
x	coordinate, m

$Greek\ letters$

σ	surface tension, N m^{-1}
μ	viscosity, Pa s
λ	eigenvalue in Kronig-Brink model and Handlos-Baron model

Supscripts

с	continuous phase
d	dispersed phase.

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