

INFLUENCES OF INTERFACIAL PHENOMENA ON THE FLUID DYNAMICS AND MASS TRANSFER OF SINGLE DROPLETS IN MICELLAR LIQUID/LIQUID SYSTEMS

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ABSTRACT

To improve reaction rates and separation processes of catalysed reactions smart solvent systems can be applied. One example for smart solvent systems are micellar liquid/liquid systems. Here, amphiphilic molecules (surfactants) are used as additives. Due to their structure these molecules will adsorb at interfaces where they influence the occurring transport processes which have a huge impact on the yield and selectivity of chemical reactions. To gain a better fundamental understanding of the occurring transport processes this work focuses on transport processes in micellar liquid/liquid systems. The results of this work show that other interfacial phenomena than “just” adsorption processes have to be taken into consideration for the description of the observed transport processes. Otherwise, the mass transport will be over estimated.

NOMENCLATURE

<i>c</i>	[mol/L]	Concentration
<i>C</i>	[-]	Drag coefficient
<i>C</i>	[-]	Counter flow cell
<i>d</i>	[m]	Diameter
<i>T</i>	[K]	Temperature
<i>t</i>	[s]	Time
<i>R</i>	[-]	Rising test cell
<i>v</i>	[m/s]	Velocity
<i>Mo</i>	[-]	Morton number
<i>Pe</i>	[-]	Pecllet number
<i>Re</i>	[-]	Reynolds number
<i>Sc</i>	[-]	Schmidt number
<i>Sh</i>	[-]	Sherwood number
<i>We</i>	[-]	Weber number
Special characters		
α	[-]	Volume fraction
β	[m/s]	Mass transfer coefficient
γ	[N/m]	Interfacial tension
μ	[Pas]	Viscosity

μ^*	[-]	Viscosity ratio
ρ	[kg/m ³]	Density
Subscripts		
<i>CMC</i>		Critical micelle concentration
<i>c</i>		Continuous phase
<i>D</i>		Dispersed phase
<i>SDS</i>		Sodium dodecyl sulfate
<i>TX-100</i>		Triton X-100

INTRODUCTION

Micellar liquid/liquid systems are examples for smart solvent systems, which can be applied to increase the reaction rates of multiphase systems and are able to improve the separation process [1-2]. Furthermore, these systems fulfil many principles of the “Green Chemistry” e.g. high and selective yields and using water as a solvent [3]. Nevertheless, more than one fluid phase occurs in these systems; hence transport processes cannot be neglected to understand the reaction mechanisms, completely [4]. Due to the presence of surfactants in micellar systems the complexity increases. Surfactants adsorb at the liquid/liquid interfaces where these molecules influence the occurring transport processes [5-6]. To quantify and to gain a fundamental understanding of the influences exerted by surfactants on the transport processes single droplets are observed in this work. With the simplification complex swarm effects have not to be taken into consideration.

The fluid dynamics of single droplets is a useful tool to quantify the interface’s characteristics which can be further used to predict mass transfer rates. Therefore, the fundamental understanding of the fluid dynamics in micellar systems is of great interest to understand the occurring transport processes. The surfactant molecules adsorb at the interface, where these molecules decrease its mobility [7-8]. At a certain surfactant concentration the droplets behave like rigid spheres. This

surfactant concentration depends on the regarded systems and surfactants. Wegener and Paschedag [5] observed toluene droplets rising in an aqueous SDS solution. For concentrations of approximately 10^{-3} mmol/L the toluene droplets behaved like rigid spheres. Whereas, a SDS concentration of 1 mmol/L reduced the velocity of tetrachlormethane droplets settling in an aqueous phase to the velocity of particles with rigid interfaces [9]. Although, the SDS concentration differs widely in both test systems, the interfacial coverage is approximately 50%. These results agree well with the calculated results by Cuenot et al. [10] and can be referred to the different adsorption behaviour in the different test systems [11]. As soon as droplets behave like rigid spheres shear stress is not transported across the interface and the inner circulations disappear. The inner circulations have a huge impact on the mass transport. Besides the additional mass transfer resistance which arises from the adsorption layer of the surfactant molecules the change of the fluid dynamics is responsible for a reduction of the mass transfer. Therefore, two limiting cases can be defined; both cases and the mechanisms responsible for the reduction of the mass transfer coefficient are schematically shown in Figure 1.

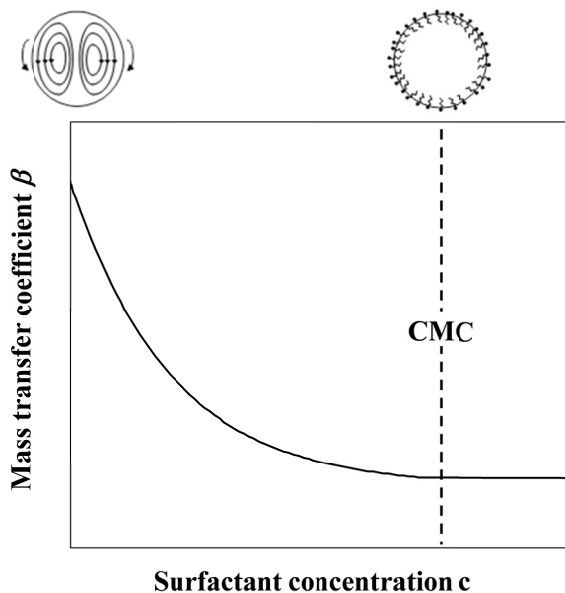


Figure 1 Mass transfer coefficient as a function of the surfactant concentration (schematically).

As schematically shown in Figure 1 and explained above the mass transfer coefficient decreases with an increase of surfactant concentration. This was observed by many authors. Nevertheless, most authors carried out their experiments at low surfactant concentrations (below the critical micelle concentration CMC). In this work the transport processes were observed in micellar liquid/liquid, respectively at high surfactant concentrations. At these concentrations the situation becomes more complex, especially for non-ionic surfactants. For these systems a change of the phase behavior must be taken in consideration besides the adsorption processes of the surfactant molecules [12].

EXPERIMENTAL SETUP

For the determination of the fluid dynamics and the mass transfer at single droplets in micellar liquid/liquid systems two different test cells were applied. Basically, the test cells were explained by Wegener et al [13] and by Paul et al. [11]. Both are shown in Figure 2. Figure 2a gives the schematic flow sheet of a 1000 mm high glass column ($d_c = 75$ mm), which is surrounded by a jacket made of acrylic glass and filled with glycerine. This jacket offers the ability to temper the setup (9a) and further ensures a good optical accessibility. Therefore, a droplet which is produced at a nozzle (6a) by a Hamilton PSD/2-module (4a, syringe pump) and loosened by a solenoid device (5a) can be tracked by a Photonfocus® MV-752-160 high-speed camera. The droplet's path is analyzed with Image-Pro Plus® 5.1 by Media Cybernetics to determine the fluid dynamics of single droplets. For the determination of the mass transfer a glass funnel (8a) is used. The glass funnel is adjustable in its height; hence different contact times can be realized. At the neck of this funnel a small amount of dispersed phase is kept; that droplets are able to coalesce. The dispersed phase is pumped out of the test cell by another syringe pump (4a). In this work an azo dye Pyridin-2-azo-dimethylaniline (PADA) was used as transferred component. Therefore, the quantification of the mass transfer was carried out by a photometer (Specord 210, Jena Analytik).

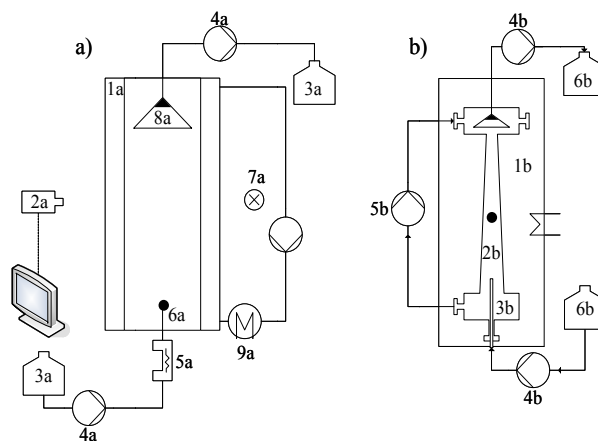


Figure 2: Experimental setups a) rising test cell: (1a) glass column with acrylic glass jacket, (2a) high speed camera, (3a) storage dispersed phase, (4a) Hamilton® PSD/2 modules, (5a) solenoid device, (6a) nozzle, (7a) illumination, (8a) glass funnel, (9a) thermostat; b) counter flow cell: (1b) heating control, (2b) glass cone, (3b) nozzle, (4b) Hamilton® PSD/3 modules, (5b) gear pump, (6b) storage dispersed phase.

The test cell shown in Figure 2a has its limitation due to its height. To observe longer contact times a second test cell was applied. The basic idea of the test cell shown in Figure 2b is the same. There is also a syringe pump (4b) for the production of the droplet. But after the droplet's release of the nozzle (3b) the droplet rises in a cone. The gear pump (5b) produces a counter flow within the cone; hence the droplet can be kept for a free selectable time.

In this work water was used as the continuous phase and 1-octanol was applied as the dispersed phase. Two different surfactants were used. SDS was used as an ionic surfactant and Triton X-100 was applied as a non-ionic surfactant. The critical micelle concentration for SDS is 8.2 mmol/L and for Triton X-100 0.2 mmol/L [11].

RESULTS: FLUID DYNAMICS

The fluid dynamics of single droplets was determined by the test cell shown in Figure 2a. The drop rise velocity is a useful tool to determine the interfacial characteristics. In presence of surfactants the interfacial coverage can be derived from these results and can be used to choose the appropriate correlation for the prediction of mass transfer rates, contact time in a column etc. A large overview of correlations for the prediction of drop rise velocities or mass transfer rates is given in the work of Wegener et al. [14]. The experimental results of the drop rise velocity shown in Figure 3 are compared with the two limiting cases described in Figure 1.

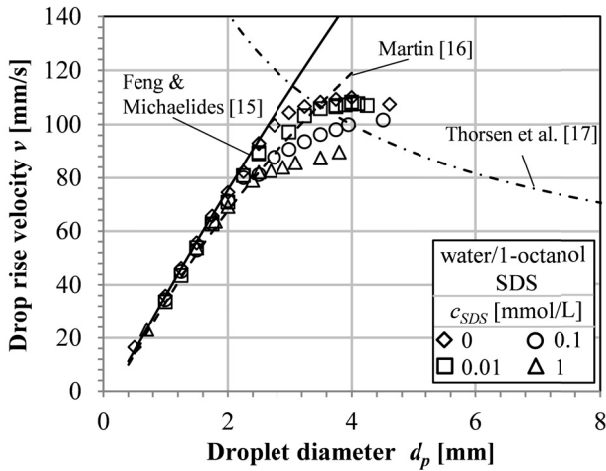


Figure 3 Drop rise velocity at steady state as a function of the droplet diameter for various SDS concentrations.

The velocity of a droplet freely rising in a continuous can be derived from the force balance [14]. The instantaneous drop rise velocity is given by:

$$\frac{dv_p}{dt} = \frac{|\Delta\rho|}{\rho_d + \alpha\rho_c} g - \frac{3}{4} C_D \frac{\rho_c}{\rho_d + \alpha\rho_c} \frac{v_p^2}{d_p} \quad (1)$$

where α represents the volume fraction of the accelerated ambient fluid by the droplet. C_D is the drag coefficient, which characterizes the interface. For the two limiting cases (free movable interface and rigid interface) the correlations for the drag coefficients by Feng and Michaelides [15] and by Martin [16] were applied to Eq. 1. After the acceleration of the droplet the droplet travels with a constant velocity through the continuous phase. The drop rise at steady state conditions is given as a function of droplet diameter in Figure 3 for various SDS concentrations. In the pure system the drop rise velocity of 1-octanol droplets rising in water is described well with Eq. 1

and the correlation for the drag coefficient by Feng and Michaelides [15]. This correlation is valid for spherical droplets. For droplets that are larger than 3 mm the spherical shape is lost and the deviations between the experimental results and the calculated values increase. The correlation by Thorsen et al. [17] can be used to calculate the rise velocity of non-spherical droplets [14]. The largest droplet produced with this setup had a diameter of 4.5 mm . Due to the interfacial tension of 8.1 mN/m [11] it was not possible to observe larger droplets.

With an increase of SDS concentration the drop rise velocity decreases. Furthermore, the droplets lose their spherical shape at lower drop diameters which can be referred to the lower interfacial tension. Both effects are shown in Figure 3. For SDS concentrations of 0.01 mmol/L the drop rise velocity hardly changed. Neither changed the droplet deformation. SDS concentrations of 0.1 mmol/L lead to an inhibition of the mobility of the droplet's interface. The drop rise velocity is decreased to the value of a particle with a rigid interface. This is well described by the drag coefficient of Martin [16]. Furthermore, the droplets deformed at smaller droplet diameters. The change in the drop rise velocity can be explained by adsorption processes at the interface. With exceeding an interfacial coverage of approximately 50% the droplets behave like rigid spheres [11]. For non-ionic surfactants the situation becomes more complex. In these systems a change of the phase behavior must be taken into consideration [18].

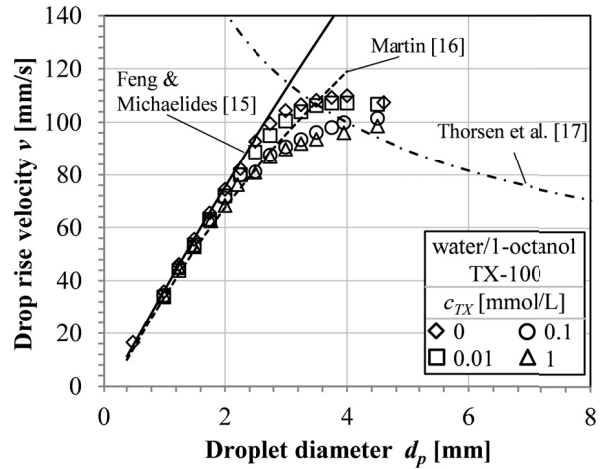


Figure 4 Drop rise velocity at steady state as a function of the droplet diameter for various Triton X-100 concentrations.

Figure 4 gives the results of the drop rise velocity at steady state for 1-octanol droplets rising in various aqueous Triton X-100 solutions. The pure system (without surfactants) is already described in Figure 3. Although, both surfactants have different adsorption behavior in the regarded test system [11], both results look similar. Below Triton X-100 concentrations of 0.01 mmol/L there was hardly any change in the drop rise velocity recognized. Exceeding Triton X-100 concentrations of 0.1 mmol/L the rigidity of the liquid/liquid interface increased and the droplets behaved like rigid spheres. This result was

unexpected due to the different adsorption behavior [11]. To compare both surfactants observed in this work the drag coefficient is given as a function of Reynolds number in Figure 5 for surfactant concentration 1 mmol/L.

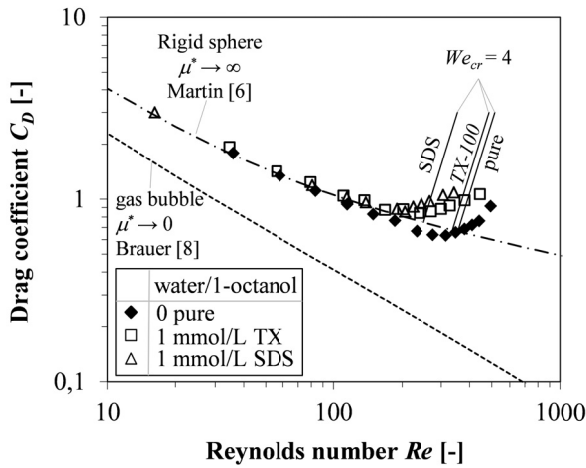


Figure 5 Drag coefficient as a function of Reynolds number for 1-octanol droplets rising in aqueous SDS and Triton X-100 solutions with similar concentrations.

Next to the experimental results the limiting cases for the drag coefficient of rigid spheres by Martin [16] and for spherical gas bubbles by Brauer [19] are given in Figure 5. Furthermore, the point of deformation is shown. According to Harper [19] the critical Weber number is given at a value of four. Therefore, the drag coefficient can be calculated by:

$$C_D = \frac{Mo \cdot Re^4}{48} \quad (2)$$

where Mo is the Morton number. In the pure test system the onset of the deformation is described well by Eq. 2. The calculated onset of the deformation is shifted towards lower Reynolds numbers with increasing surfactant concentration. This behavior is referred to the decrease of the interfacial tension. As shown in the work of Paul et al. [11] the influence of Triton X-100 on the interfacial tension is not as distinct as the influence of SDS. Therefore, the 1-octanol droplets start to deform at lower Reynolds numbers in presence of SDS than in presence of Triton X-100 (same concentration). The calculated value of the droplet deformation influenced by 1 mmol/L SDS is predicted well by Eq. 2. For Triton X-100 the largest deviations can be observed between the experimental results and the predicted value. In the spherical regime 1-octanol droplets have slightly higher drag coefficients in presence of Triton X-100 than in presence of SDS. This result is unexpected. By applying the results gained by Paul et al. [11] a change of the drop rise velocity caused by adsorption of Triton X-100 at the interface was not expected at a concentration of 1 mmol/L. For Triton X-100 concentrations of 1 mmol/L an interfacial coverage of less than 1% was calculated, hence the influence observed cannot be explained by simple adsorption process. The change of the phase behavior must be taken into

consideration. At Triton X-100 concentration higher than 0.1 mmol/L the CMC is exceeded. But this concentration is not high enough that microemulsion phases in the bulk are able to form. Nevertheless, the concentration at the liquid/liquid interface is not known and compared to the bulk phases the Triton X-100 concentration is high. In the work of Paul et al. [12] a phase diagram for the ternary system water/1-octanol/Triton X-100 is shown. The formations of microemulsions or even liquid crystalline conditions were observed in the bulk phases. These changes of the phase behavior were observed for much higher Triton X-100 concentration than in this work. But in the work of Paul et al. [12] was shown that at the interface, phases of microemulsions occur at lower concentrations than in the bulk phase. This was shown by applying the oscillating drop method which can be used to determine the interfacial rheology. With an increase of Triton X-100 concentration an increase of the visco-elastic modulus was observed [12]. The visco-elastic modulus represents the force which sets up against the deformation of a droplet. With an increase of the surfactant concentration, respectively a decrease of the interfacial tension the force needed to deform a droplet should decrease. In this work the force needed to deform a droplet was increased, which is explained by the change of the phase behavior at the interface.

Similar results were obtained in the work of Paul et al. [11]. In this work the force needed to deform a droplet's interface was determined by a colloidal probe atomic force microscopy technique. A small silica particle was used to deform a water/1-octanol interface in dependence of SDS and Triton X-100 concentration. In presence of SDS the force needed for the deformation behaved as expected. With an increase of the SDS concentration the interfacial tension decreased and the force needed to deform the interface followed. For Triton X-100 the situation was different. Here, the interfacial tension remained constant until the CMC was exceeded. The same was observed for the force needed to deform the droplet's interface. With exceeding the CMC the liquid/liquid interface became blurry and further a direct proportionality between the interfacial tension and the force needed to deform the liquid/liquid was not recognized. This effect was also referred to a change of the phase behavior at the interface.

The drop rise velocity of single droplets can be used to predict the interfacial coverage. At an interfacial coverage of approximately 50% droplets and bubbles behave like rigid spheres. This is valid as long as adsorption processes are the only processes which must be observed. For non-ionic surfactant the situation becomes more complex and is not as predictable. Spherical 1-octanol droplets can be well described by the given correlations in the literature.

Besides for the calculation of the contact time in an extraction column the fluid dynamics can be used to determine the interfacial characteristics which is useful to decide which correlation should be used for the prediction of mass transfer rates, for instance. In the regarded system water/1-octanol a SDS concentration of 0.1 mmol/L is needed to decrease the velocity to the values of a rigid sphere, the same is true for Triton X-100. Therefore, the mass transfer coefficient should

decrease until surfactant (SDS or Triton X-100) concentrations 0.1 mmol/L are set up.

RESULTS: MASS TRANSFER

The mass transfer at single droplets is observed in both test cells shown in Figure 2. The rising test cell (R) is used for short contact times and the counter flow cell (C) is applied for longer contact times. Like in the experimental investigations concerning the fluid dynamics 1-octanol droplets were observed in various aqueous solutions. The azo dye (PADA) which was used as the transferred component had a partition coefficient of approximately 60 [12]. Therefore, the mass transfer direction was chosen from the continuous phase to the dispersed phase. For SDS the measurement technique shown in Figure 2 failed. With an increase of SDS the coalescence of the droplets in the funnels was hindered. It was not possible to determine the real contact time. In presence of the non-ionic surfactant Triton X-100 the coalescence did not fail the results are shown in Figure 6.

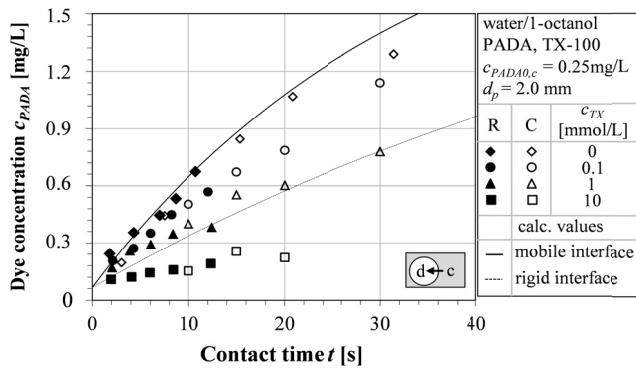


Figure 6 Comparison of the mass transfer in the system water/1-octanol for various Triton X-100 concentrations between experimental and calculated results for the limiting cases mobile interface [21] and rigid interface [21,22].

The experimental results for 2 mm 1-octanol droplets given in Figure 6 were compared with the calculated results which were predicted by correlations from the literature. Due to the high partition coefficient of PADA an external problem must be observed. The time-dependent PADA concentration was calculated by applying the correlation developed by Cliff et al. [21]:

$$c_{PADA,d}(t) = \left[1 - \exp\left(-3Fo_c \frac{Sh_c}{2 \cdot K^*}\right) \right] K^* \cdot c_{PADA,c} \quad (3)$$

The limiting cases mobile interface and rigid interface were calculated by using different Sherwood correlations. The time-dependent progress of the PADA concentration in the pure system (mobile interface) was calculated by the Sherwood correlation of Cliff et al. [21]:

$$Sh_c = f \frac{2}{\sqrt{\pi}} Pe_c^{1/2} \quad (4)$$

where f is a correction factor which takes the impact of the inner circulations into consideration. Therefore, this factor is a function of Reynolds number. For large Reynolds numbers the value of f approaches a value of one [14]. Regarding rigid spheres the inner circulations disappear. In this case the Sherwood number can be described by a correlation by Lochiel and Calderbank [22]:

$$Sh_c = 0.84 \cdot Re^{0.33} \cdot Sc^{0.5} \quad (4)$$

The exponents were derived from the boundary layer theory. By applying the Sherwood correlations to Eq. 3 it is possible to calculate the instantaneous progress of the PADA concentration for both limiting cases. The amount of PADA that is transported during the droplet formation was determined experimentally by interpolating the mass transfer for short times; hence the instantaneous progress does not start in the origin of the diagram. For the pure system the experimental results and the calculated values agree well with each other. Referring to the fluid dynamic and the physicochemical effect the mass transfer is decreased in presence of surfactants. This was observed in this work, but the amount of the reduction was unexpected. Regarding the results gained from fluid dynamics of single droplets (Figure 4) a reduction of mass transfer rates was expected up to a Triton X-100 concentration of 0.1 mmol/L. Exceeding this surfactant concentration a further reduction of the mass transfer rate was not expected. The experimental results show different behavior. Triton X-100 at a concentration of 0.1 mmol/L lead to a reduction of the mass transfer. Although, the drop rise velocity of a rigid sphere was observed (see Figure 4) the mass transfer rate is higher than the calculated progress for a rigid sphere. This is referred to Marangoni convection which is exerted by adsorption and desorption processes of Triton X-100 molecules at the liquid/liquid interface. Therefore, the mass transfer rate was not expected to be further decreased. For Triton X-100 concentrations of 1 mmol/L the mass transfer rate was further reduced and can be well described with the limiting case of a rigid sphere. For surfactant concentrations of 10 mmol/L the reduction of the mass transfer was even more distinctive. At these concentrations the mass transfer rate was even below the calculated progress for a rigid interface. For Triton X-100 concentrations above 0.1 mmol/L micelles form which agglomerate at the interface and form a microemulsion layer [11]. This layer creates an additional mass transfer resistance. As described above the formation of the microemulsion at the liquid/liquid interface was observed by the fluid dynamics (see Figure 5). Furthermore, specific experiments were carried out to characterize the liquid/liquid interface [11-12], both lead to the result that a change of phase behavior is responsible for the distinctive change of the interfacial properties with exceeding the CMC. In ternary systems consisting of water/1-octanol/Triton X-100 these phenomena must be taken into consideration. Otherwise, the mass transfer will be over predicted.

Figure 7 gives the dynamic mass transfer coefficient for various Triton X-100 concentrations to underline the impact of the phase behavior on the mass transfer. The mass transfer

coefficient is reduced with an increase of Triton X-100 concentration. There is hardly any difference between progress of the dynamic mass transfer resistance in presence of 0.1 mmol/L or 1 mmol/L. Furthermore, the mass transfer coefficient is higher than the calculated values for rigid spheres which were referred to Marangoni effects. The additional reduction at higher surfactant concentration than 1 mmol/L of the mass transfer results from the change of phase behavior at the liquid/liquid interface.

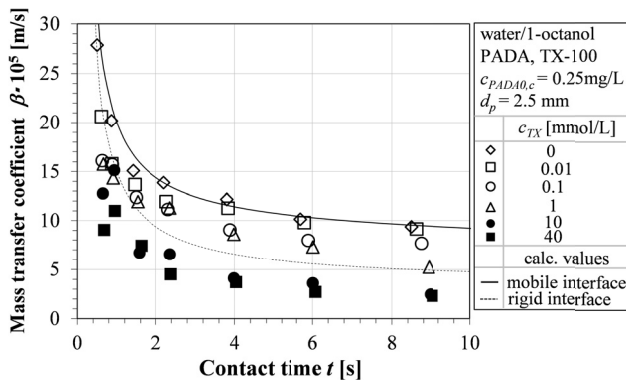


Figure 7 Dynamic mass transfer coefficient for various Triton X-100 concentrations compared with the limiting cases mobile interface [21] and rigid interface [21,22].

CONCLUSION

In this work the transport processes in micellar liquid/liquid have been observed and compared with correlations and mechanisms given in the literature. For the ionic surfactant SDS only adsorption processes at the liquid/liquid interface occur and the results observed in this work match with the expectations. For an interfacial coverage of approximately 50% the 1-octanol droplets behave like particles with rigid interfaces. For non-ionic surfactants the situation is more complex. In these ternary systems the phase behavior must be taken into consideration. Without regarding the formation of microemulsion layers at the interface the transport processes cannot be described satisfactorily. The ternary systems water/organic phase/ non-ionic surfactants are very interesting smart solvent systems, because high reaction rates and fast separation can be achieved in these systems. But the transport processes have high influences on the reaction rate [4] and are strongly influenced by surfactants as shown in this work. For the fundamental understanding of reactions in micellar liquid/liquid it is necessary to understand the interfacial phenomena as well as transport processes. Especially, the interfacial phenomena exerted by the change of phase behavior should be characterized more detailed in future (e.g. measurement of the Brewster angle).

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