Article

Study on the Mechanism of Asphaltenes Reducing Oil-Water Interfacial Tension

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As high polar components of crude oil, asphaltenes play a significant role in reducing oil-water interfacial tension(IFT). In this paper, the effects of asphaltenes on reducing IFT in the presence of surfactant were compared, and the mechanism of asphaltenes reducing the IFT was studied by the dynamic interfacial tension(DIFT) equation. Whether asphaltenes were added to the oil or 2,5-dimethyl-4-(4-dodecyl) benzene sodium sulfonate(p-S14-4) was added to the water phase, either of all results in the IFT reducing and the IFT is related to the coverage and the mass of asphaltenes adsorption at the interface. In the presence of asphaltenes, the adsorption of the active substances to the interface is not entirely dependent on diffusion, and the process can be divided into three regions. Region I: the IFT rapidly reducing, this process is controlled by diffusion of surfactant; Region II: the IFT reducing slowly, resulted from the lower diffusion rate that is limited due to the aggregates formed by the interaction of asphaltene-asphaltene; Region III: the interaction of asphaltene-asphaltene is broken by the interaction of surfactant-asphaltene. The asphaltene aggregates are reduced and adsorbed rapidly at the interface. Furthermore, the results reveal that the asphaltenes concentration affects the coverage rate and adsorption at the interface.

Keywords Asphaltene; Dynamic interfacial tension(DIFT); Synergistic interaction; Adsorption; Equilibrium interfacial tension

1 Introduction

When two different phases are in contact with each other, the interfacial tension(IFT) will be generated due to the difference in the properties of the two phases. The greater the difference between the two phases is, the larger the IFT will be. In other words, IFT is a measurement of the differences in the properties of the two phases. Usually, the difference between gas and liquid phases is expressed by the surface tension. For liquid/liquid phases, the interfacial tension is used to express the difference between the two phases[1]. Water is a polar molecular liquid, while oil(usually alkalines, etc.) is a non-polar molecular liquid. When the oil phase and water phase are in contact with each other, a transition layer——the interface layer of the oil and water phases is formed. When the surface-active substances are present, it is generally believed that the surface-active substances will spontaneously diffuse and adsorb on the interface layer to change IFT[2].

As high polar substances in crude oil, asphaltenes can also change IFT. In the laboratory, asphaltenes are defined as substances that are insoluble in *n*-alkanes(*n*-pentane, *n*-hexane, *n*-heptane, etc.) but soluble in benzene(or toluene)[3—7]. Asphaltenes are complex mixtures that are composed of aromatic hydrocarbon rings with surrounding aliphatic chains, heteroatoms, such as N, O, S, and metallic elements, such as V, Ni, Fe[8,9]. To understand the mechanism of asphaltenes on reducing the IFT of oil-water, the adsorption of asphaltenes at the interface has been studied extensively[10—15]. By MD simulation, Santos *et al.*[10] found that the adsorption mechanism of asphaltenes is not controlled by diffusion. Zhang *et al.*[11] systematically studied the oil-water interface by vertical drop method, and found that the adsorption process can be divided into three regimes along with the change of adsorption time: the decrease of dynamic interfacial tension (DIFT) of asphaltenes adsorbed on the oil-water interface is controlled by diffusion; the steric-hindrance of the adsorbed asphaltenes at the oil-water interface inhibits the further adsorption of asphaltenes to the interface; the adsorbed asphaltenes or asphaltene aggregates reconfigure, resulting in a sustain but a very slow decreasement in DIFT. This study provided a further understanding of the adsorption mechanism of asphaltenes. Mohammadi *et al.*[12] found that only asphaltene monomers participate in the adsorption process by estimating the short-term equation of Ward-Tordai and the diffusion coefficient. Jian *et al.*[13] studied the effect of the concentration of asphaltenes on the IFT of oil-water by using the hanging drop method and molecular dynamics simulation and found that in toluene solution, all asphaltene molecules indeed migrate to the interface, but not all asphaltene molecules can migrate into the interface, which was consistent with the experimental study of Mohammadi *et al.*[14]. Moreover, Mohammadi *et al.*[14] and Campen *et al.*[15] respectively proposed new models to study the adsorption of asphaltenes. The new model was reported by Mohammadi *et al.*[14]. They combined the model with the Frumkin/Langmuir isotherm to estimate the apparent diffusion coefficient and

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adsorption kinetics of asphaltenes with different concentrations, and the results showed that the diffusion migration of asphaltenes to the interface of oil-water decreased with increasing the asphaltenes concentration. Camben *et al.*[15] proposed an adsorption reaction-diffusion model for asphaltenes, which described the adsorption process of asphaltenes from toluene solution to the gold surface as three parallel processes: volume equilibrium between different aggregates; diffusion of aggregates to the solid surface; surface adsorption. Although the model proposed by Campen[15] is related to the solid phase, there was a certain similarity with the results of the study by Zhang *et al.*[11]. It was also confirmed that the asphaltenes adsorption is not controlled by diffusion, which was also consistent with the results of Santos *et al.*[10].

Although previous studies have all proved that the adsorption of asphaltenes to the interface is not a simple diffusion process, the presence of asphaltenes has an effect on the IFT of oil-water. It still has certain limitations. At present, in the stage of tertiary oil recovery[16], the chemical flooding method of a ternary composite displacement fluid(alkali+ surfactant+polymer)[17] is usually used for oil recovery. When the IFT of oil-water reaches the ultra-low level, the oil recovery will be significantly improved[18]. Therefore, it is necessary to investigate the effects of different asphaltenes on the interfacial tensions(IFTs) of surfactant solutions. Moreover, asphaltenes obtained from different sources, different solvents, and different physical and chemical environments are different[4]. Therefore, this paper compared the efficiency of asphaltenes in reducing IFT of oil-water. And studied the adsorption mechanism of asphaltenes and the effect of asphaltenes concentration on IFT in the presence of salt(NaCl) and surfactant[2,5-dimethyl-4-(4-dodecyl) benzene sodium sulfonate, p-S14-4] in an aqueous solution.

2 Experimental

2.1 Materials

The surfactant p-S14-4 was synthesized in our laboratory. The asphaltenes were extracted from Daqing Crude Oil(China) by the SARA(Saturate, Aromatic, Resin, Asphaltene) separation method. Toluene and sodium chloride were purchased from the Beijing Chemical Plant(China). All reagents were analytical, and the water was deionized.

2.2 Methods

2.2.1 Spinning Drop Tensionmetry

The asphaltene toluene solutions of different mass concentrations were used as the oil phase. The aqueous phase was an aqueous solution containing 1.00%(mass ratio) NaCl and 0.10%(mass ratio) p-S14-4. The DIFT was measured by the Spinning drop method. The temperature was 45℃, the oil phase was 0.8 μL, and the revolution was 5000 r/min. The calculation equation of IFT was as follows:

$$\gamma = 1.2336 \times 10^{6} \cdot \frac{\Delta\rho \cdot D^{3}}{P^{2} \cdot n} \cdot f(L/D) \qquad (1)$$

where γ is the interfacial tension(mN·m^{-1}), $\Delta\rho$ is the density difference between the oil phase and aqueous phase, D is the diameter of oil drop(cm), L is the length of oil drop(cm), P is the speed reciprocal(ms·rev^{-1}), n is the refractive index of aqueous phase(generally n=1.330), $f(L/D)$ is the correlation factor[when $L/D \geqslant 4$, $f(L/D)$=1].

2.2.2 Asphaltenes Adsorption Experiment

According to the Ward-Tordai's short-time approximation [equation (2)], the short time DIFT[equation (3)], and long time DIFT[equation (4)], when the concentration of surfactant in solution is kept constant, and the adsorption and diffusion were controlled, (γ_0–γ_t) has a linear relationship with $t^{0.5}$ and γ_t has a linear relationship with $t^{-0.5}$, the diffusion coefficient(D_{ap}) can be estimated from the slope[19]. In this experiment, the concentrations of surfactant(0.10%, mass fraction) and sodium chloride(1.00%, mass fraction) in the aqueous phase remained constant, the concentration of asphaltenes was changed. The change of IFT was monitored, and the adsorption of asphaltenes at the oil-water interface at different concentrations was compared.

$$\Gamma_t = 2\,(D_{\mathrm{ap}}/\pi)^{0.5} c t^{0.5} \qquad (2)$$

$$(\gamma_0 - \gamma_t)/c = 2RT(D_{\mathrm{ap}}/\pi)^{0.5} t^{0.5} \qquad (3)$$

$$t \xrightarrow{r_t} \infty = \gamma_{\mathrm{e}} + n(RT\Gamma^{2}/c)(7\pi/12D_{\mathrm{ap}}t)^{0.5} \qquad (4)$$

where n is a constant, γ_{e} is interfacial tension in infinite time.

3 Results and Discussion

3.1 Efficiency of IFT Reduction by Asphaltenes

First of all, the influence of the changed composition of oil and water phases on the dynamic interfacial tension(DIFT) was measured. The composition of different oil-water systems is listed in Table 1, and the results are shown in Fig.S1(see the Electronic Supplementary Material of this paper). The results show that the composition of the water phase and oil phase indeed influences IFT. The addition of asphaltenes to the oil phase or the addition of surfactant to the water phase

Table 1 The composition of different oil-water systems

System	Constituent
①	Toluene+NaCl
②	Toluene+NaCl+p-S14-4
③	Toluene+NaCl+asphaltenes
④	Toluene+NaCl+p-S14-4+asphaltenes

significantly reduces the equilibrium interfacial tension($\mathrm{IFT_{eq}}$), but the efficiency of reducing IFT is different. The addition of asphaltenes results in a decreased value of 14—18 mN/m. The addition of p-S14-4 results in a decreased value of 20.58 mN/m.

Because p-S14-4 is a highly polar anionic surfactant composed of hydrophilic and hydrophobic groups, asphaltenes, formed by the interaction of dipoles-dipoles, can be regarded as surfactant with certain polarity. Therefore, compared with asphaltenes, p-S14-4 has a stronger ability to reduce the IFT. Based on the addition of NaCl to the water phase, the influence of various factors on the reduction of oil-water IFT is listed in Table 2.

Table 2 Comparison of effects of aqueous phase and oil phase composition changed on $\mathrm{IFT_{eq}}$

Mass concentration of asphaltenes(%)	$\mathrm{IFT_{eq}}$/(mN·m^{-1})				
	①–②	①–③	①–④	③–④	④–②
3.00	20.85	17.53	19.17	1.64	1.68
1.50	20.85	16.38	21.25	4.86	0.39
0.50	20.85	14.93	21.48	6.55	0.62
0.25	20.85	16.03	21.59	7.75	0.73
0.05	20.85	15.03	21.42	6.39	0.57

When p-S14-4 surfactant was not added to the aqueous phase, the asphaltenes can be better dissolved in toluene due to certain solvation between toluene and asphaltenes[20]. Therefore, the addition of asphaltenes to the toluene oil phase brings about a decreased value of 14—18 mN/m, indicating that polar asphaltenes have a certain interface activity. The reduction of IFT caused by asphaltenes addition has two main contributions. Firstly, the addition of asphaltenes changes the properties of the toluene oil phase. It increases the polarity of the oil phase and reduces the polarity difference between the oil and water phase, which is the main factor for the reduction of IFT. Secondly, the adsorption of asphaltenes as active substances at the interface results in the reduction of IFT, which makes a small contribution. The reason is that asphaltenes mainly exist in the form of aggregates in toluene[21], its diffusion rate is slow, and its adsorption at the interface is difficult. This can be seen from the influence of asphaltenes concentration on IFT. The lower the asphaltenes concentration is, the smaller the aggregates will be, and asphaltenes with small molecules are more conducive to its diffusion to the interface and adsorption. However, when the asphaltenes concentration decreases from 3.00%(mass fraction) to 0.05% (mass fraction), the decreased value of IFT varies from 17.53 mN/m to 15.03 mN/m, indicating that the higher the asphaltenes concentration is, the stronger the ability to reduce IFT will be. It can be seen that in the system of NaCl aqueous phase-toluene oil phase, the adsorption of asphaltenes to the interface has little influence on IFT. In other words, the presence of asphaltenes at this time mainly affects the polarity of the toluene oil phase, while the influence of water on the asphaltene molecules at the interface layer is limited.

When p-S14-4 surfactant was added to the aqueous phase, the IFT of the oil-water decreases further when asphaltenes were added to the system of the p-S14-4+NaCl aqueous phase-toluene oil phase, and the decreased value is greater than that when only p-S14-4 was added to the water phase(except asphaltene 3.00%, mass fraction). In other words, the addition of p-S14-4 to the water phase and asphaltenes to the oil phase are conducive to the reduction of oil-water IFT, that is, the synergistic interaction between p-S14-4 and asphaltenes at the interface is positive. When asphaltenes concentration is high(3.00%, mass fraction), asphaltene molecules near the interface can also interact with p-S14-4 molecules. However, due to the high asphaltenes concentration, it is not enough to depolymerize asphaltene aggregates and the viscosity of the oil phase is high, so the effect of changing the properties of the interface layer is weak. The IFT value is greater than that of the pure toluene oil phase when the concentration is 3.00%, the value of system ④–② is 1.68 mN/m. However, when the asphaltenes concentration is less than 3.00%, the strong interaction between p-S14-4 and asphaltenes leads to the depolymerization of aggregates of asphaltene at the interface, forming small molecules of asphaltene and adsorbing on the interface. Together with p-S14-4, a relatively stable adsorption layer is formed. Therefore, IFT shows a considerable decrease.

Secondly, it was found that the interaction between p-S14-4 and asphaltenes is related to the concentration of asphaltenes. Within a certain concentration range, in the system of p-S14-4+NaCl water-oil phase, the lower the concentration of asphaltenes is, the better the reduction of oil-water IFT will be. As can be seen from Table 2(①–④), when asphaltenes concentration decreases from 3.00% to 0.25%, the decreased value of IFT varies from 19.17 mN/m to 21.59 mN/m. When asphaltenes concentration is 0.05%, the decreased value of IFT is 21.42 mN/m, slightly less than that when asphaltenes concentration is 0.25%, and this decreased value is closed to the decreased value of 20.85 mN/m when p-S14-4 was added in the aqueous phase alone. This may be because the concentration of asphaltenes at this time is too low, and the amount of asphaltenes that interacted with p-S14-4 at the interface is small, so it has little influence on the oil-water interface, and this influence may be related to the arrangement at the interface.

In conclusion, the addition of asphaltenes in the oil phase alone or surfactant in the water phase alone will reduce IFT. The asphaltenes and p-S14-4 can interact synergistically at the interface, enhancing the ability to reduce IFT. And within a certain concentration range, the lower the asphaltenes concentration is, the stronger the synergistic interactions with

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p-S14-4 will be.

3.2 Analysis of the Arrangement of Asphaltenes at the Interface

As mentioned above, the influence of asphaltenes on oil-water IFT is related to the distribution of asphaltenes in the interface layer. Moreover, some studies showed that IFT depends on the surface coverage[22], rather than the time. When the asphaltenes coverage of the oil-water interface reaches 35%—40%, the adsorption rate of asphaltenes at the interface will decrease[11], which will have an impact on IFT. Therefore, the coverage rates of asphaltenes with different mass concentrations at the oil-water interface were compared in this experiment. According to the equation

$$\ln\left(\frac{\mathrm{d}\pi}{\mathrm{d}t}\right)=\ln(b)-\frac{\pi\Delta A}{K_{\mathrm{B}}T}\tag{5}$$

where $\pi=\gamma_0-\gamma_t$, the equation can be simplified as

$$\ln\left(\frac{\mathrm{d}\pi}{dt}\right)=\ln(b)-\frac{\gamma_0\Delta A}{K_{\mathrm{B}}T}+\frac{\gamma_t\Delta A}{K_{\mathrm{B}}T}\tag{6}$$

$$\ln\left(\frac{\mathrm{d}\pi}{\mathrm{d}t}\right)=B+\frac{\gamma_t\Delta A}{K_{\mathrm{B}}T}\tag{7}$$

Plotted with $\ln(\mathrm{d}\pi/\mathrm{d}t)\sim\gamma_t$(as shown in Fig.1), we can compare ΔA of the asphaltenes covering the area at the interface from its slope. The higher the slope is, the greater the ΔA will be.

As shown in Table 3, the smaller the asphaltenes concentration is, the larger the area occupied by the asphaltenes at the interface(except 3.00%, mass fraction) will be. This is because asphaltenes diffuse faster to the interface of oil-water at a lower concentration, so the amounts of asphaltenes adsorbing at the interface increase. Because asphaltenes can be adsorbed at the interface between toluene and water, the polyaromatic nuclei of asphaltene molecules tend to extend parallel to the interface. When the asphaltenes mass concentration is 3.00%, due to its higher concentration, asphaltenes mostly exist in the form of aggregates, and its size is larger, so the diffusion and adsorption rates are small. However, as large aggregates, once it is adsorbed into the interface, it covers a large area of the interface.

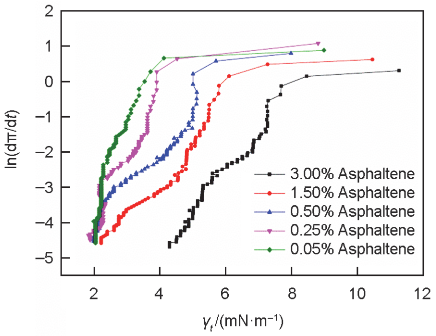

Fig.1 Comparison of ln(d*π*/d*t*) *vs.* *γ*$_t$ with different mass concentrations of asphaltenes

Table 3 The comparison of Δ*A*/*K*$_{\mathrm{B}}$*T* of different mass concentrations of asphaltenes

Mass concentration of asphaltenes(%)	$\Delta A/(K_{\mathrm{B}}T)$
3.00	1.17
1.50	1.01
0.50	1.03
0.25	1.75
0.05	2.96

By compared the relationship between IFT$_{\mathrm{eq}}$ of oil-water and asphaltenes concentration(as shown in Fig.2), the results show that when the asphaltenes mass concentration is 0.25%, the IFT$_{\mathrm{eq}}$ is the smallest(less than the IFT$_{\mathrm{eq}}$ when the asphaltenes mass concentration is 0.05%), which might be due to the fact that the content of asphaltenes monomer is the largest at the interface in this concentration, its distribution at the interface and the synergistic interaction of asphaltenes and surfactant are better. Therefore, the change of IFT is not only related to the coverage rate of asphaltenes but also related to the adsorption amount at the interface and the synergistic interactions with the surfactant.

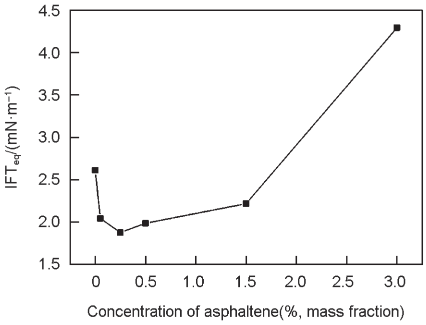

Fig.2 Curve of equilibrium interfacial tension with different asphaltenes concentrations

3.3 Adsorption of Asphaltenes to Oil-water Interface

As mentioned above, the synergistic interaction of asphaltenes and surfactants results in the reduction of oil-water IFT, which may be related to the adsorption of asphaltenes to the interface. Therefore, to explore the mechanism of the reduction of IFT under the synergistic interaction of asphaltenes-surfactant, the DIFT of the system of toluene-water phase with different asphaltenes concentrations was carefully measured. According to the Ward-Tordai equation, short-time DIFT equation and long-time DIFT equation were used to compare the relationship curves between (γ_0–γ_t) and $t^{-0.5}$ and between γ_t and $t^{-0.5}$, respectively. The results are shown in Fig.3. As can be seen in Fig.3, all relationship curves can be divided into three regions: Region I, Region II, and Region III. As shown in Fig.3(A), in Region I of the short-time DIFT equation curve, the presence of asphaltenes has little effect on the change of its

DIFT. The calculated apparent diffusion coefficient is roughly the same as that of the simple structure surfactant(as shown in Table 4), which indicates that in the system of p-S14-4+NaCl water-toluene oil phase, the adsorption of active substances at the interface in the initial short time of DIFT is controlled by the diffusion of surfactant. In the presence of asphaltenes, the IFT is greater than that in the absence of asphaltenes in a short period. This result can be interpreted as that D_{ap} in the absence of asphaltenes is greater than that in the presence of asphaltenes, indicating that the presence of asphaltenes will slow down the diffusion of surfactant p-S14-4, resulting in less adsorption amount of the interfacial layer per unit time.

Similarly, Region II and Region III are stages that are closed to IFT_{eq} of the long-term DIFT equation curve in Fig.3(B). It is found that in Region II, regardless of the presence of asphaltenes, the relation of IFT and $t^{0.5}$ is linear. The slopes in the presence of asphaltenes at this stage are smaller than those in Region I. Therefore, the diffusion process of asphaltenes to the oil-water interface at this stage is very slow, because asphaltene aggregates are formed by the interaction of asphaltene-asphaltene.

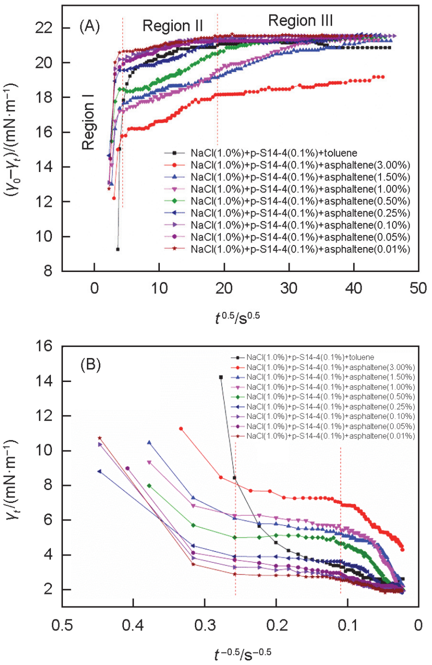

Fig.3 Comparison of dynamic interfacial tension between pure toluene and toluene solutions of asphaltenes
(A) Short-time DIFT curves; (B) long-time DIFT curves.

Table 4 Apparent diffusion coefficients(D_{ap}) of Region I at different asphaltenes concentrations

Mass concentration of asphaltenes(%)	$10^7 D_{ap}$
0	83.50
0.01	11.00
0.05	8.34
0.10	8.92
0.25	3.83
0.50	3.50

In Region III, in the absence of asphaltenes, IFT rapidly reaches IFT_{eq} after experiencing Region I and Region II. In the presence of asphaltenes, the rate of IFT reaching IFT_{eq} is relatively slow. It can be understood that in the absence of asphaltenes, due to the rapid diffusion of the surfactant, it is easy to reach the adsorption equilibrium. However, in the presence of asphaltenes, after Region II, asphaltenes break through the energy barrier, making more asphaltenes adsorb on the oil-water interface. The interactions of the asphaltene-water phase and asphaltene-surfactant enhance, and are the aggregated asphaltene molecules depolymerize and are quickly adsorbed at the interface. Therefore, the IFT of oil-water is influenced by the depolymerization speed of asphaltene aggregates, which show that the IFT of oil-water decreases continuously until the final IFT_{eq} reaching. These processes of three regions are shown in Fig.4.

To sum up, it can be inferred the interface adsorption mechanism of the system of the p-S14-4+NaCl water-toluene oil phase. Region I: the D_{ap} of this stage is roughly the same as that of the surfactant, and this stage is controlled by the diffusion of surfactant. Region II: in this stage, asphaltenes slowly diffuse and are adsorbed toward the interface. And the diffusion rate is limited due to the aggregation formed by the interaction of asphaltene-asphaltene. Region III: the interaction of asphaltene-asphaltene is broken, the interactions of surfactant-asphaltene and water-asphaltene are enhanced, the particle size of the asphaltene aggregates decreases, this stage is controlled by asphaltenes depolymerization rapid adsorption at the interface.

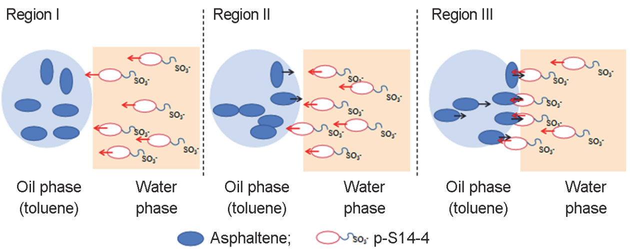

Fig.4 Process of asphaltenes and surfactants in different regions

3.4 Interfacial Arrangement of Asphaltenes

As mentioned above, asphaltenes have a certain influence on the properties of the oil-water interface. The influence is related to the asphaltenes concentration, the interaction of surfactant-asphaltene and asphaltene-water, and the rate of adsorption, etc. When the asphaltenes concentration is 3.00%, it usually exists as aggregates and the adsorption to the interface is slow, thus its distribution at the interface and the interaction with surfactants are the lowest. However, when the mass concentration is 0.50%—1.50%, the interaction between asphaltene molecules becomes weakened, with its adsorption to the interface increasing, so the distribution at the interface and its interaction with surfactants at the interface is increased. When the asphaltene concentration is 0.25%, the adsorption to the interface becomes faster, the interaction of asphaltene-asphaltene is the weakest, therefore the distribution of asphaltene at the interface and the interaction with surfactant is the strongest. Therefore, the distribution of asphaltenes at the interface was simply inferred as shown in Fig.5.

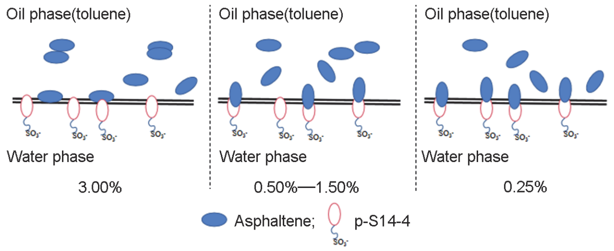

Fig.5 Distribution of asphaltenes at the oil-water interface with different concentrations

4 Conclusions

In the system of NaCl aqueous-toluene oil phase, no matter the addition of asphaltenes in toluene alone or the addition of surfactant p-S14-4 in the aqueous phase alone, either of which results in IFT reducing. The addition of asphaltenes results in a decreased value of 14—18 mN/m. The addition of p-S14-4 results in a decreased value of 20.58 mN/m. At the same time, the addition of p-S14-4 to the water phase and asphaltenes to the oil phase further reduce the IFT, asphaltenes and p-S14-4 have a synergistic interaction at the interface. The change of IFT is not only related to the coverage rate of asphaltenes at the interface but also related to the adsorption mass at the interface and the synergistic interaction with the surfactant. In the presence of asphaltenes, the process of adsorption is not controlled by diffusion, and this process can be divided into three regions. Region I：the IFT rapidly reducing, this process is controlled by diffusion of surfactant；Region II：the asphaltenes are adsorbed slowly toward the interface, and the diffusion rate is limited due to the aggregates formed by the interaction of asphaltene-asphaltene, the IFT reducing slowly; Region III：the interaction of asphaltene-asphaltene is broken, the interactions of surfactant-asphaltene and water-asphaltene enhance. The asphaltene aggregates particle size decrease, rapid adsorb in the interface that the adsorption speed is controlled by asphaltenes depolymerization. Moreover, the ability of asphaltenes on IFT reducing and its adsorption to the interface are both influenced by the concentration. In a certain concentration range, the lower the asphaltenes concentration is, the faster the adsorption on the interface and the smaller the IFT of oil-water would be.

Electronic Supplementary Material

Supplementary material is available in the online version of this article at http://dx.doi.org/10.1007/s40242-021-1178-3.

Conflicts of Interest

The authors declare no conflicts of interest.

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