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Original article

Study on the mechanism of surfactant flooding: Effect of betaine structure

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Abstract:
In order to elucidate the oil displacement mechanism of micro-emulsions formed by different betaines at pore throats, this study selected three betaine surfactants with different hydrophobic branched chains for a microscopic visualization oil displacement experiment. The interfacial tension, dilational modulus, interactions of oil droplets, and apparent viscosity of the emulsions were measured. Besides, the microscopic oil displacement mechanism and oil displacement effects of different betaines in homogeneous and heterogeneous models were investigated. The results revealed the beneficial interfacial activity and viscosity enhancement effects of the three betaine solutions. With the increase in the branched degree of betaines, the strength of interfacial films and the viscosity enhancement effect decreases. In the homogeneous model, betaine solutions emulsify crude oil into droplets with strong interfacial films. The in-situ plugging effect improves oil recovery and the sweep efficiency in the pore throats, and the remaining oil is mainly in the form of droplets. As the branched degree increases, the strength of the interfacial films and the oil recovery decline. In the heterogeneous model, the plugging effect enhances the pore structure heterogeneity. The three betaine solutions can increase the sweep efficiency but the displacement solutions only migrate along the dominant pathway within the sweep range. As a result, a large amount of isolated cluster residual oil remains, resulting in similar oil recovery efficiency for betaine flooding to that of water flooding in the heterogeneous model.

1. Introduction

With the increasing demand for petroleum resources, mid-high permeability reservoirs have entered the middle-late period of exploitation. Insufficient remaining crude oil results in difficulty in extraction (Feng et al., 2018; Shao et al., 2023). Although low-permeability reservoirs are abundant, the small pore throats and complex structures lead to problems such as low recovery efficiency, strong heterogeneity and injection difficulty (Feng et al., 2018; Li et al., 2022). Therefore, it is of great significance to improve the recovery method and enhance the residual oil recovery by using surfactants after water flooding in low-permeability reservoirs (Kamal et al., 2017).

Surfactant flooding is a high-efficiency approach to enhance oil recovery and has a wide range of applications (Zhao et al., 2022b). The mechanism of surfactant flooding activates the residual oil in reservoir pores by interfacial tension (IFT) reduction, emulsification, and wettability alteration (Arabloo et al., 2016a; Kumar and Mandal, 2020). The reduction in interfacial tension can increase the number of capillaries, reduce the mobilization energy of crude oil, and promote the flow of residual oil trapped in the porous media. As a result, residual oil saturation is drastically decreased (Yang et al., 2021; Sukee et al., 2022). Emulsification can produce oil-in-water (O/W) and water-in-oil (W/O) emulsions. The latter are prone to increasing the sweep coefficient by plugging and entrainment effects (Dong et al., 2012; Zhao et al., 2022a). Meanwhile,

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the mechanism for O/W emulsions is directly related to the size of oil droplets and the structure of pore throats (Kumar et al., 2021). Wettability alteration is an effective means to mobilize residual oil. Besides, the mobilization and migration behavior of crude oil is quite different under diverse wetting conditions. Changing the wettability of solid surfaces from oleophilic or hydrophilic to neutral is more favorable for the mobilization of crude oil, which will be transported away after the wettability transformation (Liu and Wang, 2020, 2022).

Considering flooding using polymers in oilfields, they can increase the viscosity, reduce the mobility ratio of crude oil to the oil displacement agent, and improve the sweep coefficient (Sidiq et al., 2019; Wang et al., 2019). However, due to the limitation of displacement efficiency, the oil recovery efficiency increase is relatively low (Browne et al., 2020). Surfactant-polymer flooding systems (Panthi et al., 2020) contain surfactants and polymers and can improve the sweep efficiency and oil washing efficiency simultaneously, which in turn substantially improves the oil recovery efficiency. Nonetheless, it is difficult to inject polymer solutions with high molecular weight into low-permeability reservoirs. Thus, viscosity enhancement by in-situ emulsification via surfactants needs to be considered (Sun et al., 2020a). Betaine is a zwitterionic surfactant that contains both positive and negative charges in its polar group. It possesses excellent properties such as foaming ability (Gao et al., 2017), low toxicity (Zhou et al., 2014), high-temperature tolerance, and good resistance to salt (Sun et al., 2020c). When betaine molecules adsorb at the oil-water interface, they have strong interactions with the active fractions of crude oil. As a result, in-situ emulsification, viscosity enhancement and sweep efficiency improvement can be achieved by regulating the strength of interfacial films (Zhang et al., 2021; Sun et al., 2023c). Unfortunately, the published reports about betaines in this field are mainly mixed solutions with other types of surfactants (Zhou et al., 2022; Ren et al., 2023), while few reports involve the emulsification regulation mechanism of single betaines.

As a novel method in the petroleum industry, microscopic visualization oil displacement experiments have been widely used to investigate the interactions of oil-brine-rock, the permeability of multiphase flow in porous media, and the mechanism of enhanced oil recovery (Zou et al., 2018; Gaol et al., 2020; Xu et al., 2020). These experiments feature good observability and can realize similar conditions to the reservoirs. Additionally, microfluidic technology presents the advantages of visualization, convenience and rapid screening ability (Conn et al., 2014; Yun et al., 2017; Sun et al., 2021). Using microfluidic models to simulate the properties and structures of rocks in the reservoir can help explore the effect of different reservoir pores on the oil recovery efficiency, which is conducive to the screening of the oil displacement systems (Rezaei Dehshibi et al., 2019). Mohammadi et al. investigated the oil displacement mechanism of multiple shale systems by spot glass micromodels with small-scale discontinuous flow barriers. By comparing the oil displacement results of polymers on homogeneous and heterogeneous models, it was found that the discontinuous flow barriers increase the sweep efficiency of polymers in the heterogeneous model and enhance the oil recovery efficiency (Mohammadi et al., 2013). Liu et al. designed a micromodel with triangular pore throats to explore the pore-scale oil recovery process of low-salinity water flooding. They demonstrated that the emulsions were more likely to coalesce to form large plugs under low salinity conditions to improve the sweep efficiency (Liu et al., 2021).

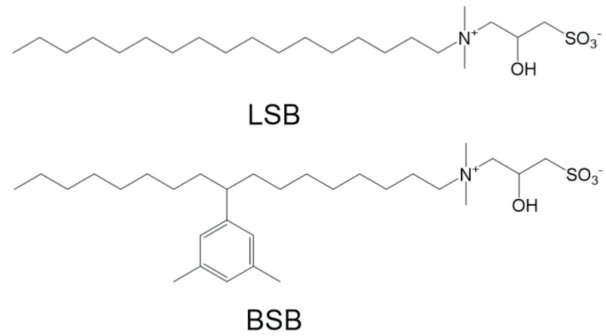

Fig. 1. Structure and abbreviation of LSB and BSB.

The interfacial properties of betaines are excellent as they show mixed adsorption with the active fractions of crude oil at the interface (Cao et al., 2016; Zhou et al., 2016; Liu et al., 2023). The size of the hydrophilic and hydrophobic groups has a decisive impact on the performance of betaine molecules (Zhang et al., 2017). In this work, oil-water IFT, the strength of interfacial films, drop coalescence, and the viscosity enhancement of emulsions are investigated to explore the influence of the branched hydrophobic groups on the properties of betaine molecules. In addition, the displacement effects of betaines with different structures are studied in homogeneous and heterogeneous microfluidic models. The oil displacement mechanism of betaine and the effect of molecular structures are obtained from the macroscopic interfacial properties and the microscopic seepage characteristics, such as the deformation and migration of crude oil in the microfluidic model. The results of this work offer theoretical guidance for the design of oil displacement using betaines.

2. Materials and methods

2.1 Materials

The zwitterionic surfactant betaines (LSB, MSB, and BSB) used in this study were obtained from the China Petroleum Exploration and Development Research Institute. The structures and abbreviations of three kinds of betaine are shown in Fig. 1. Among them, LSB has a linear chain and BSB possesses a branched alkyl chain. MSB is a mixed system of LSB and BSB, in which the mixing ratio of LSB and BSB is 1:1. Double-distilled water (resistivity > 18.2 MΩ/cm) was used in the preparation of the surfactant solution. The crude oil, obtained from PetroChina Changqing Oilfield, had a viscosity of 22.30 cP at 25 °C and 4.36 cP at reservoir temperature (63 °C). The composition of the simulated formation water is shown in Table 1. The displacement solutions were all prepared by the simulated formation water with a concentration of 0.2 wt% without special considerations.

Chip models obtained from China University of Petroleum (Beijing) were used in the microscopic visualization oil dis-

Table 1. Formulation and composition of the simulated formation water.

$CaCl_2$ (g/L)	$MgCl_2 \cdot 6H_2O$ (g/L)	Na_2SO_4 (g/L)	Na_2CO_3 (g/L)	$NaHCO_3$ (g/L)	NaCl (g/L)
0.0554	0.25412	3.9954	0	0.1776	1.3544

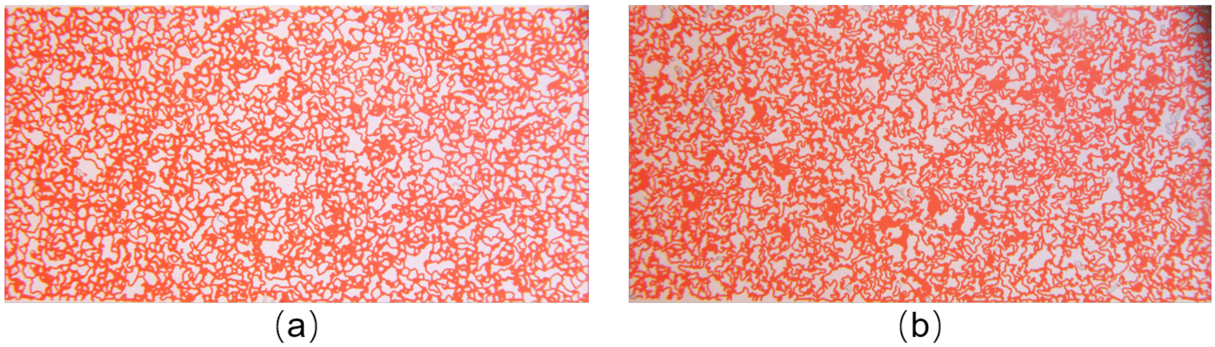

Fig. 2. Simulation model after saturated crude oil. Homogeneous (a) and heterogeneous model (b).

Table 2. Structural parameters of the two models.

Model	Porosity	Homogeneity coefficient	Pore structure factor	Throat (μm)	Pore (μm)
A	0.45	0.51	0.041	60-70	250-350
B	0.44	0.40	0.044	70-80	250-400

placement experiments. They included homogeneous and heterogeneous models and all of them were water-wet models. The structural parameters of the models after crude oil saturation are shown in Fig. 2 and Table 2.

In Table 2, porosity refers to the ratio of volume of all pores to their total volume in the model, which is equal to the red oil area/total area in Table 2 in quantity. The homogeneity coefficient is the average pore throat radius/maximum pore throat radius, with a value between 0 and 1. The larger the value, the stronger the homogeneity of the pores. Throat and pore were obtained by microscope camera capture combined with Matlab statistics, with the latter used to describe the width of the throat and the size of the pore. The pore structure factor was taken to evaluate the pore structure. The larger the B_z value, the better the seepage capacity of the reservoir. The calculation formula is:

$$B_z = \frac{V_z \overline{r}_z}{\lambda} \tag{1}$$

where $\overline{r}_z$ represents the average value of the main flow pore throat radius, λ represents the tortuosity of the pore throat, and V_z represents the total volume of the pore throat corresponding to r_z.

2.2 Apparatus and method

2.2.1 Interfacial tension test

A spinning drop interfacial tensiometer (TX-500C) was utilized to determine the dynamic interfacial tension between oil displacement solutions and crude oil (Zhou et al., 2014). The rotation speed was 5000 r/min, the temperature was 25 °C, and the concentration of oil displacement solutions was 0.2%. Crude oil acted as inner phase and the surfactant solution as outer phase.

2.2.2 Dilational rheological property test

The experiments were carried out using a drop profile analysis technique and a dynamic contact angle machine (LSA100, Eastern-Dataphy Instruments) (Zhou et al., 2014). Periodical oscillations of droplets were performed, and changes in the area were captured by a camera. The dynamic interfacial dilational properties were determined using the drop profile analysis method. The oscillating frequency was 0.1 Hz, and the volume of droplets was 8 μL (Sun et al., 2023a; Zhang et al., 2008).

2.2.3 Droplet coalescence test

A restructured interfacial tensiometer (DCAT21, Dataphysics, Germany) was employed to conduct the droplet coalescence test (Liu et al., 2019). In the experiment, upper and lower crude oil droplets (1 μL) were injected by a syringe pump, and the two oil droplets were aligned. The crude oil droplet coalescence tests were initiated after 120 seconds of aging time. The lower droplet gradually approached the upper droplet at a speed of 10 μm/s, and the force curves of the two droplets were measured using a micro-force balance.

2.2.4 Emulsion viscosity test

The crude oil and water phase were added into a tube at a volume ratio of 1:1. Subsequently, 100 times of shaking was carried out to ensure sufficient emulsification. The upper phase

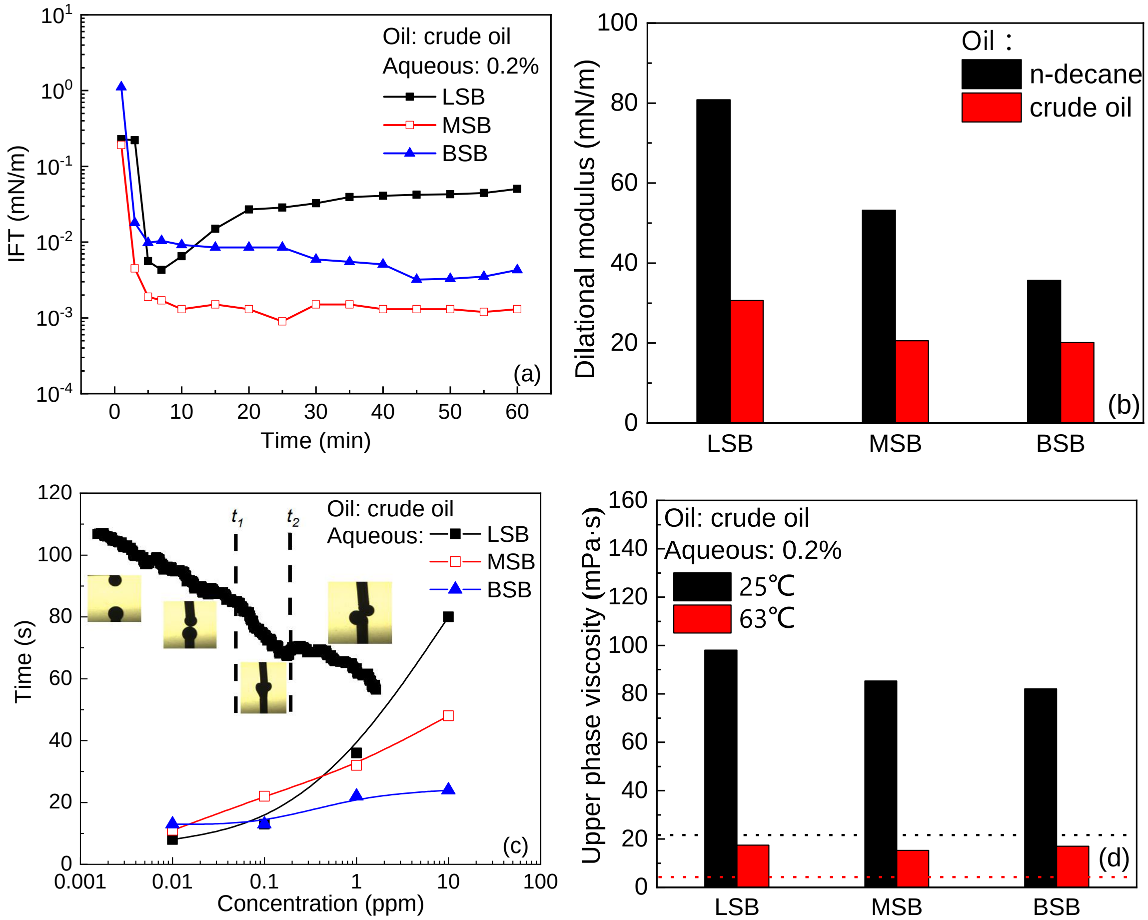

Fig. 3. (a) Dynamic interfacial tensions of three betaine solutions against crude oil, (b) comparison of interfacial dilational moduli at the water-decane and water-crude oil interfaces, (c) coalescence times of crude oil droplets in the aqueous phase as a function of betaine concentration (the inner diagram is a representative force curve in the process of droplet coalescence) and (d) the viscosity of upper W/O emulsion of three betaine solutions with crude oil at different temperatures (the dotted line indicates the viscosity of crude oil at the corresponding temperature).

viscosity of the emulsion was measured at 25 °C and 63 °C immediately by a viscometer (DV3TLVCP).

2.2.5 Microscopic visualization oil displacement experiment

The microscopic displacement experiments were conducted in a chip model with simulated pore structures (Fig. 2). The experimental schematic was consistent with the description by Mejia et al. (2019). Crude oil was first saturated into the model to ensure that it completely filled the model. Then, the displacement agents were injected by a microfluidic injection pump (FLOW-EZ S/N: 12490) at a constant injection rate of 0.1 µL/min from the right inlet and displaced from the left outlet. The displacement process was stopped when the oil production of the remaining oil in the model no longer changed. All experiments were conducted at 25 °C. The whole crude oil displacement process was recorded by a microscope camera system (Nikon SMZ18) at 1.5× magnification (Liu et al., 2019). The pressure at the inlet of the chip was recorded by a sensor of the injection pump and uProcess software during the displacement process. The pressure point at the chip outlet was connected to the atmosphere and this pressure was not recorded.

The oil displacement effect analysis was based on image capture during the displacement process. The oil displacement efficiency and sweep efficiency were calculated from the pixel data in Matlab statistics. The oil displacement efficiency was measured as the pixel points of the displaced oil/all the saturated crude oil in the chip. The sweep efficiency is the numerical difference between all crude oil and un-swept crude oil pixels/all crude oil saturated in the chip. In order to determine the experimental error, five experiments were carried out on the same standard system. The errors of oil displacement efficiency and sweep efficiency were 1% and 2% respectively.

3. Results and discussion

3.1 Interface properties

Fig. 3(a) shows the dynamic IFTs of three betaine solutions with different branched hydrophobic chains against crude oil. It can be seen that the equilibrium IFT value of LSB is only

10^{-2} mN/m order of magnitude. Notably, the equilibrium IFT values of MSB and BSB solutions can reach an ultra-low level (10^{-3} mN/m). The results demonstrate that the three different betaines possess good interfacial activity to reduce the interfacial tension. In general, the crude oil from PetroChina Changqing Oilfield has low viscosity and the size of its active components is relatively small. Therefore, only betaine with an appropriate branched chain can match the size compatibility with the active components in crude oil to achieve an ultra-low IFT (Sun et al., 2020b, 2023c). Cao et al. (2016) also reported that betaines can realize size compatibility with the active components in crude oil to form a mixed adsorption film. The synergistic effect enables surfactants to adsorb at the oil/water interface more tightly, thus decreasing the interfacial tension to the ultra-low level. In particular, the curve of interfacial tension of LSB presents an obvious "V" shape. At the oil-water interface, LSB molecules are adsorbed and their hydrophilic heads gradually lay flat at the interface. Meanwhile, the active components in crude oil also diffuse to the interface and insert into the vacancies of the LSB molecules. This results in the formation of a compacted interfacial film, and the obvious decline of interfacial tension. However, when the hydrophilic heads of the LSB molecules lie flat completely at the interface, a large interfacial space exists onat the oil side. Thus, the active components in crude oil cannot form a compacted adsorption film with the LSB molecules and leads to the increased value of interfacial tension.

The interfacial dilational modulus of the three betaines against decane/crude oil is shown in Fig. 3(b) As the hydrophilic groups of the surfactants at the interface respond to the area variation through orientation changes, the molecular rearrangements of betaines dominate and elastic films with high modulus appear (Zhang et al., 2014). The modulus of the solutions follows an order of LSB $>$ MSB $>$ BSB. Notably, the interfacial dilational modulus of LSB against decane is up to 80 mN/m. Besides, the interfacial dilational modulus values of MSB and BSB against decane are about 50 mN/m and 30 mN/m, respectively. As the branched degree of betaines increases, steric hindrance results in less orientation change of the hydrophilic groups, and diffusion exchange gradually plays a dominant role. When the surfactant molecules are squeezed out in the compression process, the strength of the interfacial film decreases (Guo et al., 2021). Due to the insertion of active components of crude oil, the modulus values of betaines against crude oil are lower than those against decane. However, LSB with a linear structure still possesses the highest modulus values. The strength of interfacial films of betaines originates from the large hydrophilic groups that lie flat at the interface. Moreover, the strength of the interfacial film decreases as the branched degree of betaines increases.

In this work, the approaching, squeezing and coalescence processes of oil droplets were characterized and the corresponding force curves were recorded in Fig. 3(c). Firstly, it can be seen that the lower droplet gradually approaches the upper droplet and the solution is squeezed out. Then, the two oil droplets begin to contact. On account of the interfacial film with a certain strength formed by the surfactants adsorbed at the oil-water interface, the two oil droplets will not coalesce immediately, but then will squeeze each other and finally coalesce. If the interfacial film is robust, the coalescence process may not manifest during the experiment. With the increase in the concentration of surfactants, the squeeze time of the two oil droplets extends. The squeeze time is the longest at a concentration of 10 ppm, which indicates that the adsorption amount of the interfacial surfactants increases gradually with increasing concentration and the growing strength of the interfacial film makes the coalescence more difficult. In the experiment, LSB with a linear chain is difficult to coalesce due to its robust interfacial film. The strength of the interfacial film for MSB is slightly lower than that of LSB. Due to this relatively weak interfacial film, BSB possesses a short squeeze time, which is consistent with the mechanism of oil mobilization in advanced water flooding, as obtained by Ayirala et al. Therein, it was confirmed that the modulus was directly related to the coalescence process of oil droplets; the lower the modulus value, the easier the coalescence process. When the small oil bank formed after coalescence, the migration of crude oil in the pores was easier (Ayirala et al., 2018).

The viscosities of W/O emulsions formed by the three betaine solutions with crude oil were measured at 25 °C and the reservoir temperature (63 °C), and the results are shown in Fig. 3(d). It can be seen that the three betaines have good emulsifying ability. In addition, the viscosities of the emulsions are all above 80 mPa·s at 25 °C, which is nearly three times that of crude oil. Therefore, the three betaines have a good viscosity enhancement effect (Kang et al., 2019). By comparison, the viscosity of the emulsion formed by LSB with crude oil is the highest. This phenomenon may be attributed to the strong interfacial adsorption film formed by betaine and the active components of crude oil. The high elasticity of the dispersed droplets results in a high apparent viscosity (Sun et al., 2023b). Similarly, the viscosity enhancement effect can be observed at 63 °C.

3.2 Oil displacement experiment on the homogeneous model

The oil displacement effect of the three betaines on the homogeneous model is shown in Fig. 4. Due to the relatively low viscosity of crude oil, the oil displacement efficiency of water flooding (69.4%) is comparatively high. It can be observed that the oil displacement efficiency of LSB, MSB, and BSB are 83.7%, 79.0%, and 74.7%, respectively, under the same condition. By comparison, the oil displacement efficiency of LSB is higher. With the increase in the branched degree of betaine, the oil displacement efficiency of the betaine solutions decreases.

The sweep effect and sweep efficiency of three betaine solutions on the homogeneous model are shown in Figs. 5(a)-5(b). Compared with water flooding, the three betaine solutions can significantly increase the sweep efficiency. It can be found that the sweep range of LSB can extend to the edge of the model, while the sweep range of MSB and BSB is similar and significantly smaller than that of LSB. Meanwhile, LSB with the highest interfacial tension has a better sweep effect than MSB and BSB, which demonstrates that the oil displace-

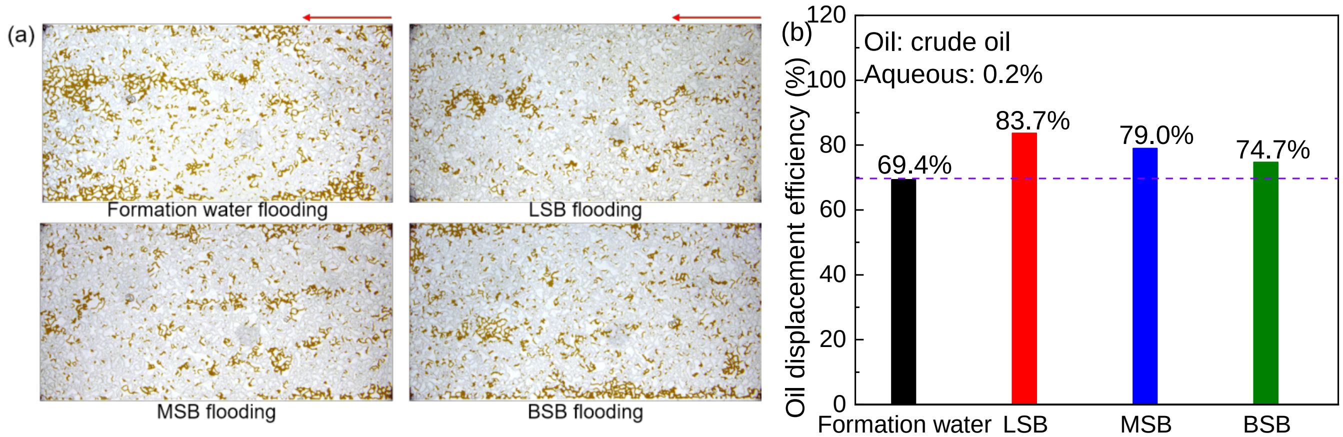

Fig. 4. (a) Oil displacement effects and (b) oil displacement efficiency of three betaine solutions on a homogeneous model. The red arrow represents the flow direction of the microfluidics.

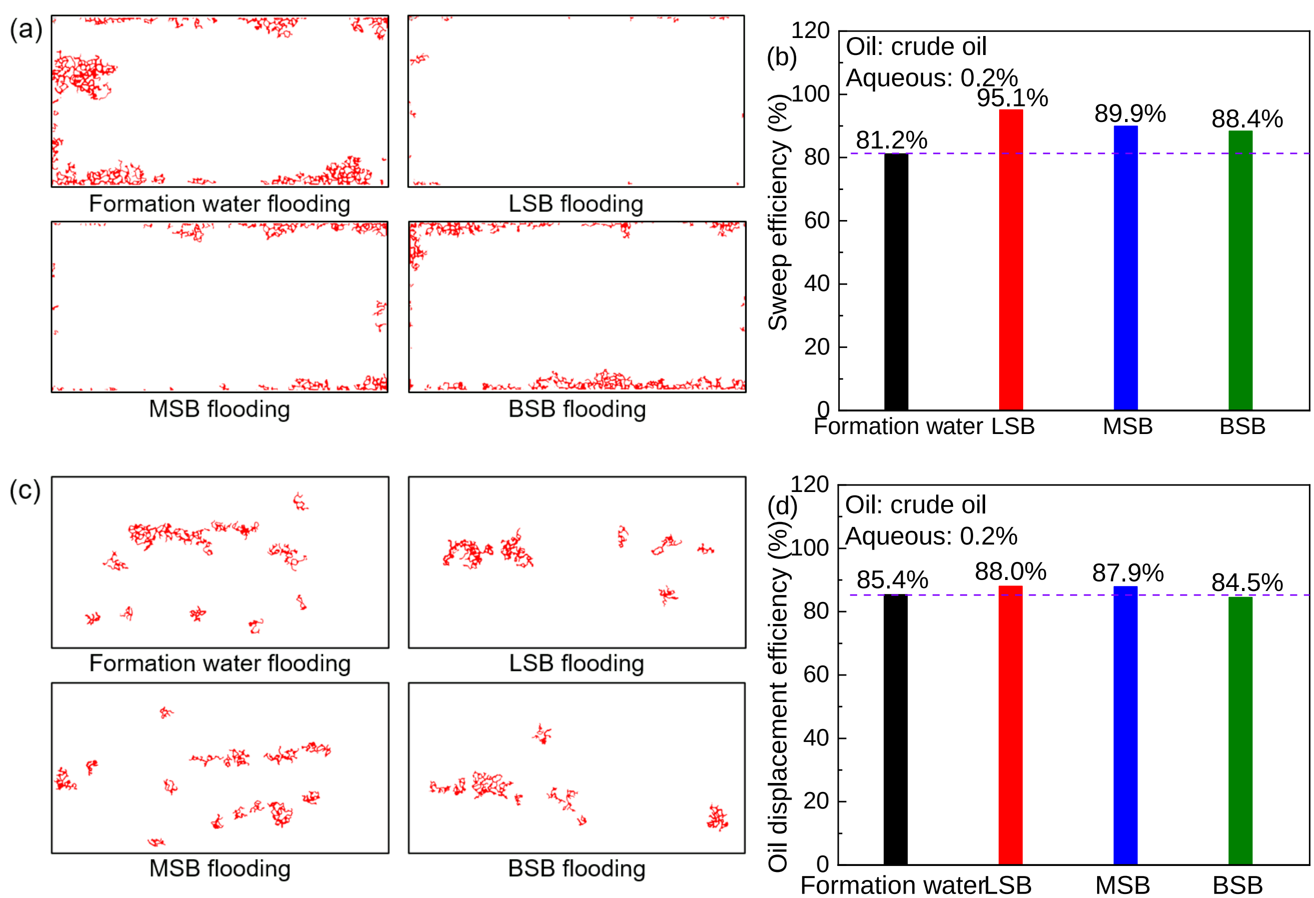

Fig. 5. (a) Sweep effects and (b) sweep efficiency of three betaine solutions on the homogeneous model and (c) the cluster remaining oil and (d) oil displacement efficiency in the sweep range of three betaine solutions on the homogeneous model.

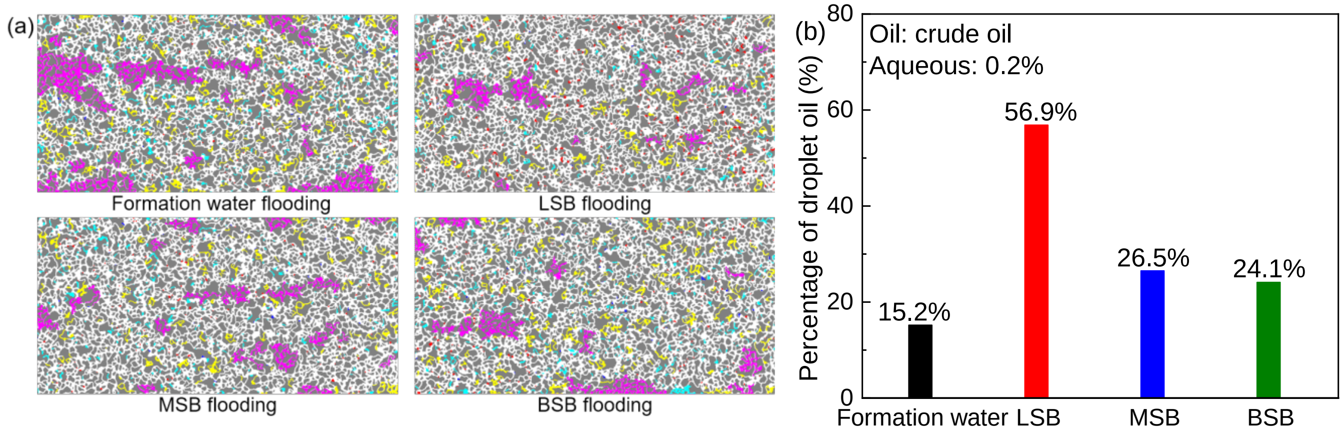

Fig. 6. (a) The classification of remaining oil and (b) proportion of dispersed O/W emulsion droplets after three betaine solutions flooding.

ment effect is not only determined by interfacial tension. The most crucial factors that determine the crude oil displacement effect also include the strength of the interfacial film. Thus, the formation water breaks through the crude oil in the model and stops sweeping after forming a dominant channel in water flooding. In contrast, due to the strong interfacial film of betaines, a blocking effect is generated by the emulsions. The betaine solutions flow in the vertical direction, resulting in higher sweep efficiency than water flooding. Importantly, the blocking effect decreases as the branched degree of betaines increases. LSB presents the highest modulus value and the best viscosity enhancement effect, and LSB is difficult to coalesce, which endows LSB with the best blocking effect. The emulsification of oil displacement agents enables the solution to possess higher viscosity and a better sweep effect, which is conducive to enhanced oil recovery.

The cluster remaining oil and oil displacement efficiency of the three betaine solutions in the sweep range on the homogeneous model are depicted in Figs. 5(c)-5(d). In the sweep range, there is little difference between the three betaine solutions in the number and area of remaining oil. Similarly, there is little difference between the three betaine solutions in their oil displacement efficiency. This proves that it is the higher sweep range of LSB that enables LSB to possess slightly higher oil displacement efficiency compared to MSB and BSB.

Fig. 6 depicts the classification of remaining oil and the proportion of dispersed O/W emulsion droplets after flooding with the three betaine solutions. The classification of residual oil types and the specific meanings represented by different colors are in good agreement with those described by Sun et al. (2023b). Specifically, the red color represents residual oil droplets. Oil-in-water emulsion droplets are all produced in the three betaine solutions and dropwise residual oil is dominant in the residual oil. Due to the synergistic effect between the betaine and the active components of crude oil, a strong interfacial film is formed under high water-cut conditions. Thus, numerous highly elastic dispersed droplets are further formed. By counting the remaining oil after the flooding with water and the three betaines, it is found that the number of oil-in-water emulsion droplets formed by oil displacement agents and crude oil is significantly higher than that of water flooding during the displacement process. Moreover, there are quite a few oil droplets trapped at the entrance of the pore throats. The number of oil-in-water droplets after LSB flooding is the highest, which also implies that the oil droplets in LSB solution are harder to coalesce than the oil droplets in MSB and BSB solutions.

On the one hand, this migration behavior can reduce the residual oil and increase the washing efficiency by producing the Jamin effect, to further increase the sweep efficiency when the droplets pass through the throat. Meanwhile, the Jamin effect is simultaneously determined by the strength of interfacial film and the interfacial tension. The area of oil droplet increases when the oil droplet deforms. In addition, the strength of interfacial films determines the difficulty of the deformation of oil droplets. The higher the strength of the interfacial films, the more difficult it is for the oil droplets to deform. Therefore, the higher the interfacial tension and the strength of interfacial films, the stronger the Jamin effect will be.

The blocking effect of O/W emulsion droplets generated during the migration of the three flooding betaine solutions is shown in Fig. 7. During the displacement process of LSB, the residual oil in the pores generates low interfacial tension to increase the capillary number and the oil droplets are mobilized to flow along the channels. Owing to the plugging effect produced by the strong interaction of interfacial films, the mobilized residual oil may be trapped by the fine channels after migrating for some distance, thus increasing the sweep efficiency.

Therefore, in the homogeneous model, the strength of interfacial film has a greater influence on the oil displacement effect than the interfacial tension. By comparison, the LSB with the strongest interfacial film has a superior oil displacement effect, higher sweep range, and better oil displacement efficiency within the sweep range.

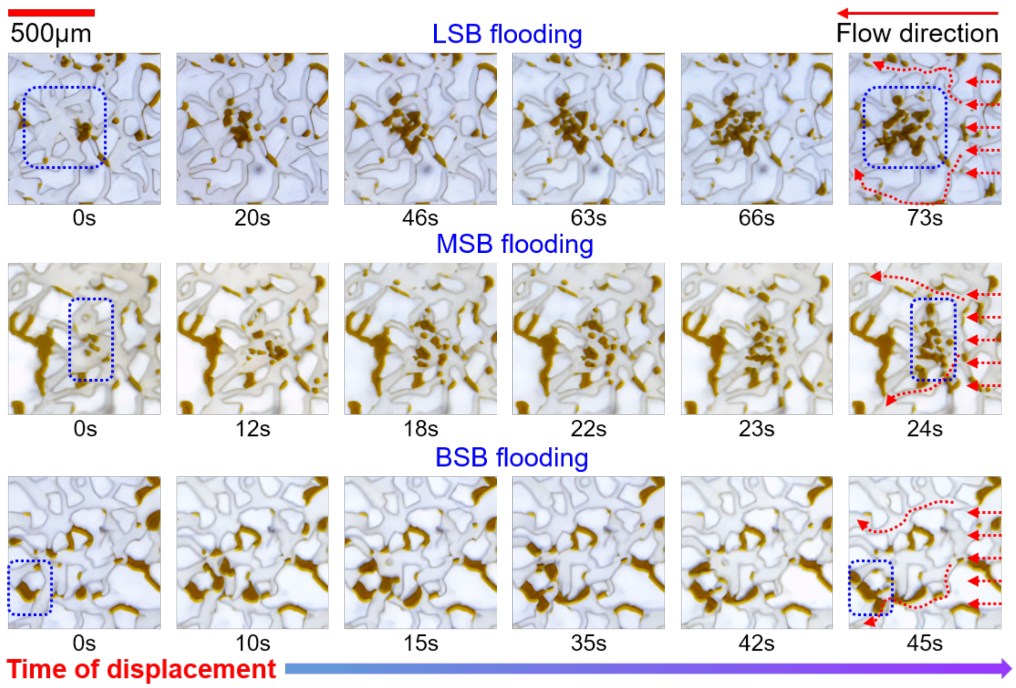

Fig. 7. The blocking effect of O/W emulsion droplets generated during the migration of three betaine solutions flooding.

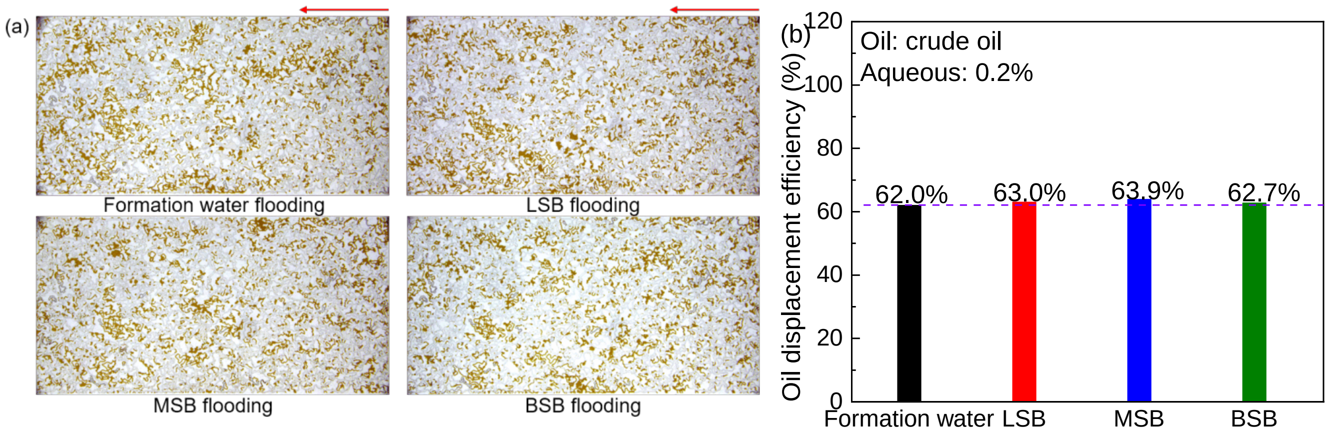

Fig. 8. (a) Oil displacement effects and (b) oil displacement efficiency of three betaine solutions on the heterogeneous model.

3.3 Oil displacement experiment on the heterogeneous model

Fig. 8 illustrates the oil displacement effects and oil displacement efficiency of the three betaine solutions on the heterogeneous model. It can be seen that the oil displacement efficiency of flooding with water and the three betaines are all approximately 62.0% ~ 64.0%. Thus, there is little difference in the oil displacement efficiency by flooding between water and the three betaines. However, it is quite different from the oil displacement effect of the three betaine solutions on the homogeneous model. Although the betaine solutions can reduce the interfacial tension to the ultra-low level, they cannot significantly enhance oil recovery in the heterogeneous model. This may be attributed to the interfacial tension reduction that leads to an easier breakthrough of the displacement solution in the heterogeneous model. After the breakthrough of the displacement solution, the bypassing phenomenon occurs and results in the reduction in oil washing efficiency. To verify this speculation, the sweep effect and the oil recovery within the sweep range were analyzed, and the results are shown in Fig. 9.

As shown in Fig. 9, the sweep efficiency of the three betaine solutions is higher than that of water flooding. This indicates that the oil droplets, which are larger than the pore throats, are captured during the migration of emulsions in the particularly heterogeneous area. As a result, the permeability of the aqueous phase decreases, which weakens the fingering and crossflow phenomenon and improves the sweep efficiency. Thus, profile modification is achieved by the emulsification and viscosity enhancement of betaine solutions, which results in a better sweep effect for flooding with betaine solutions

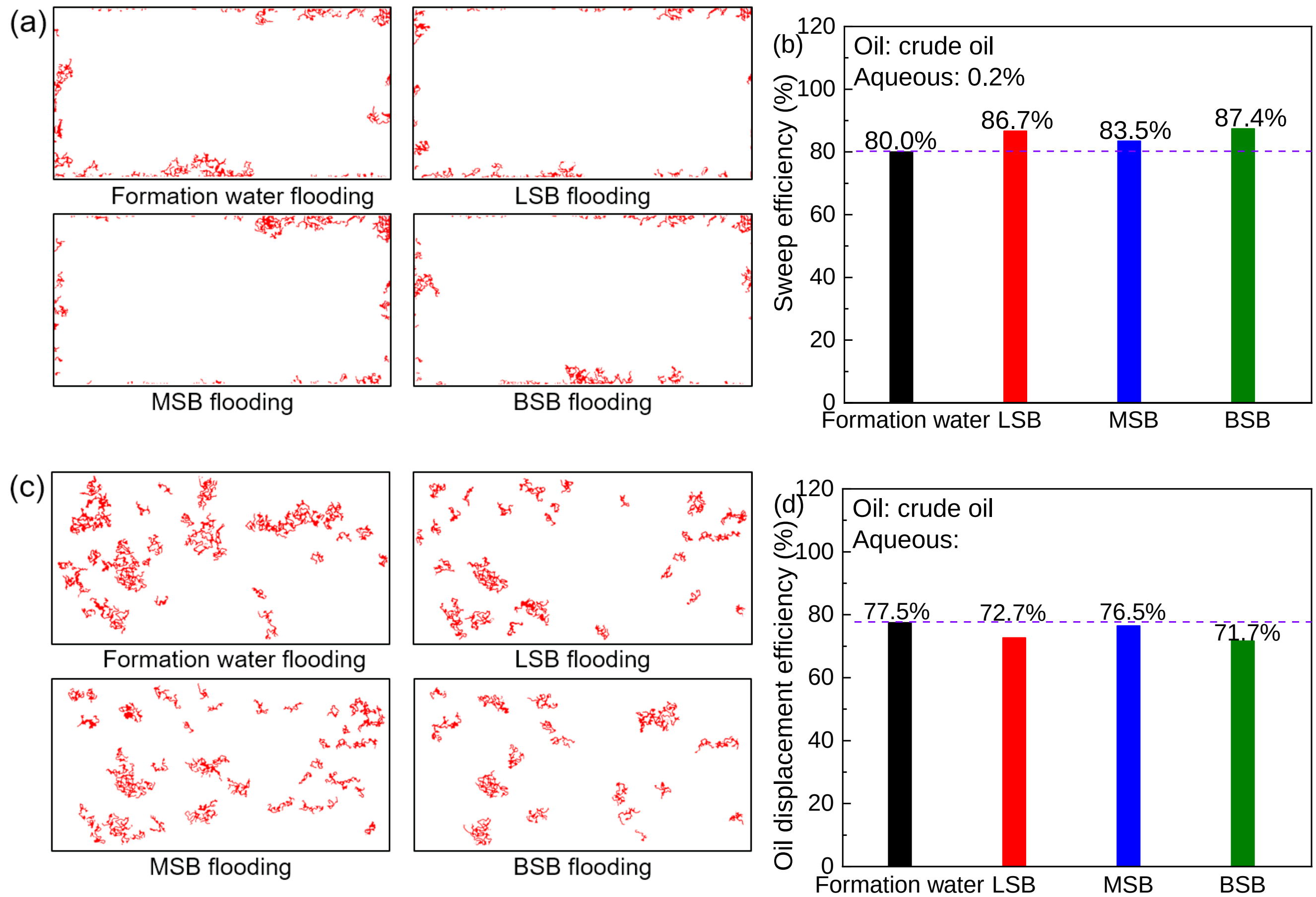

Fig. 9. (a) Sweep effects and (b) sweep efficiency of three betaine solutions on the homogeneous model and (c) cluster remaining oil and (d) oil displacement efficiency in the sweep range of three betaine solutions on the heterogeneous model.

than water.

However, the residual oil after flooding with the three betaine solutions is mainly cluster residual oil on the heterogeneous model, and the oil displacement efficiency within the sweep range decreases. Due to the heterogeneity of the model, the formation water preferentially flows along the high-permeability zone. Thus, a large area of cluster remaining oil remains after water flooding. Despite the poor sweep effect, the residual oil can still be transported as a whole within the sweep range. Compared with water flooding, the betaine flooding migrates more uniformly in the heterogeneous area.

High oil washing efficiency can be achieved due to the low interfacial tension of betaine solutions, but the viscosity enhancement and plugging effects of the emulsions cause the betaine solutions to bypass the heterogeneous areas. Thus, cluster residual oil is left in an island pattern. Therefore, the oil displacement efficiency of betaine solutions in the sweep range is slightly lower than that of water flooding. In other words, when the homogeneity of the model deteriorates, the plugging effect of betaine solutions will enhance the "local heterogeneity". The increase in sweep efficiency precisely counteracts the decrease in oil washing efficiency, resulting in little change in the overall oil displacement efficiency.

Fig. 10 shows the cluster residual oil in the sweep range after flooding with the three betaine solutions on the heterogeneous model. It is visible that the amount of cluster remaining oil after flooding with betaine solutions is more than that of water flooding. There are more cluster residual oils with small areas after flooding with betaine solutions. It is widely believed that heterogeneity tends to result in the crossflow phenomenon (Wang et al., 2022). Since the plugging effect enhances the heterogeneity of pore throats, the displacement solutions are more likely to migrate along the high-permeability pathway and the effects of the displacement surfactants become weakened.

The oil displacement effect and migration mode of the three flooding betaine solutions on the heterogeneous model are shown in Fig. 11. In the detailed figures, it can be found that the throat entrances of the cluster remaining oil after flooding with betaine solutions (blue dotted line) are all perpendicular to the mainstream displacement direction of the model (red arrow). In the weak shear area, the displacement solution can only migrate along the dominant pathway in the high-permeability zone, and the cluster remaining oil is bypassed (Li et al., 2018). This conforms with the oil displacement effect, sweep efficiency and displacement efficiency within the sweep range obtained above.

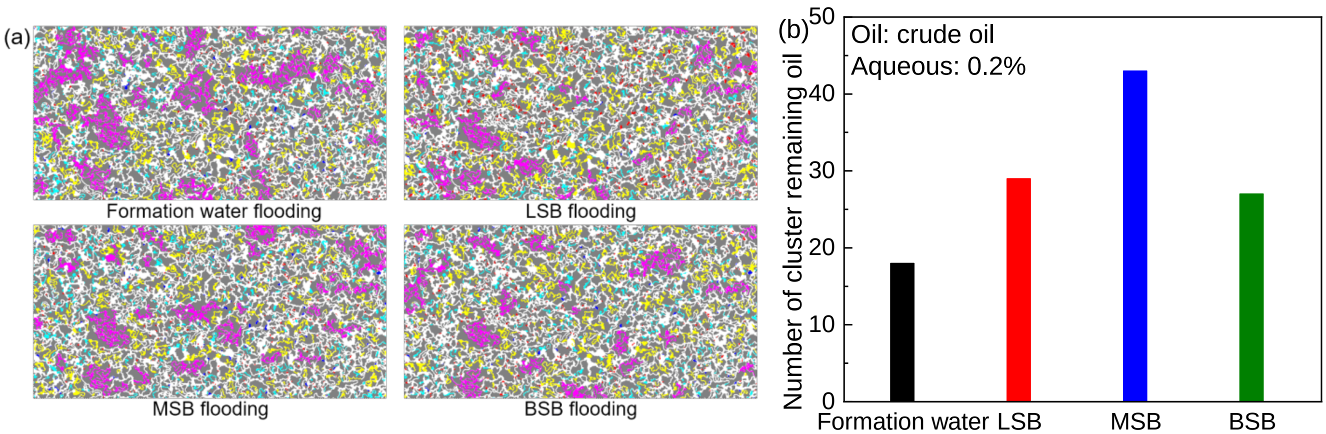

Fig. 10. (a) Classification of remaining oil and (b) number of cluster remaining oil in the sweep range after flooding with three betaine solutions on the heterogeneous model.

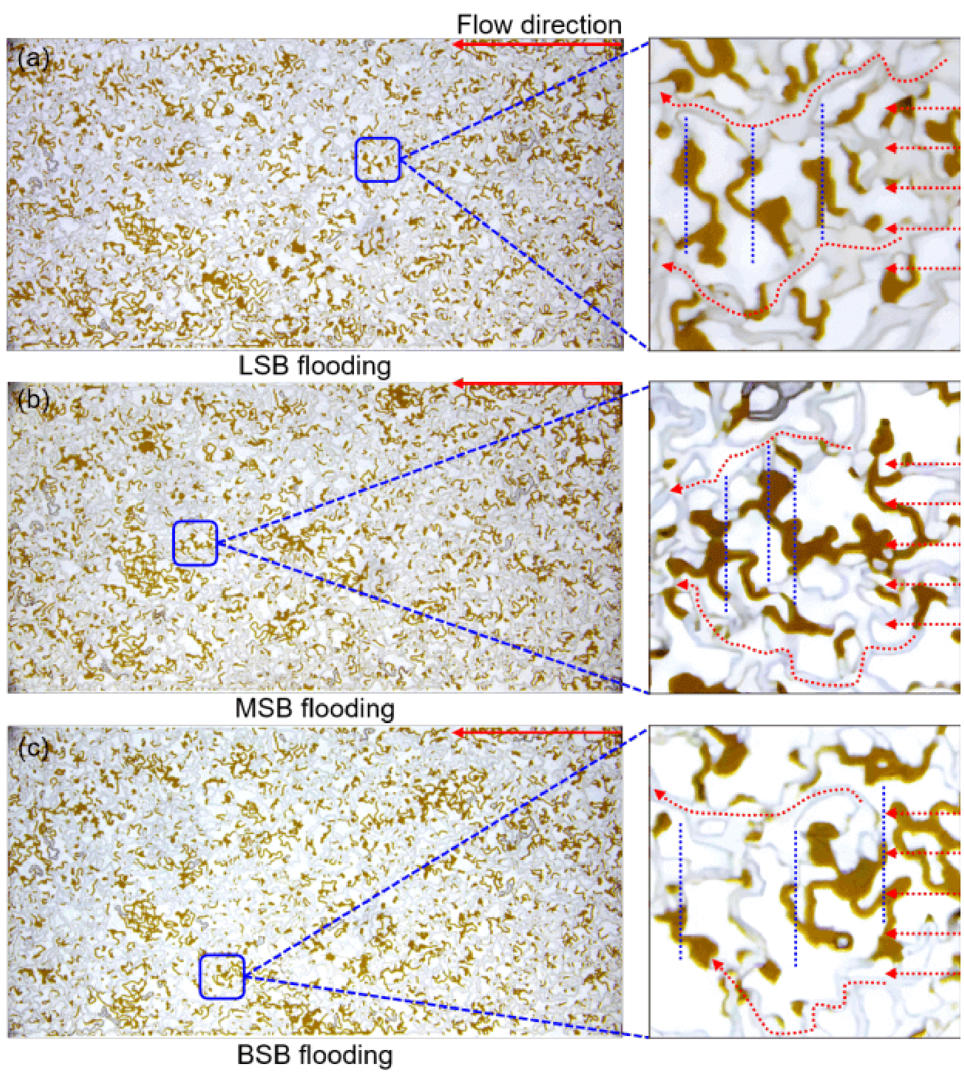

Fig. 11. Oil displacement effect and migration mode of three betaine solutions flooding on heterogeneous model, (a) LSB, (b) MSB and (c) BSB. The red arrow represents the flow direction of the microfluidics. The blue arrow represents the direction of the remaining oil in the throat.

4. Conclusions

In this work, based on the different characteristics of betaines with different hydrophobic branched chains, the oil displacement effects of three betaine solutions were obtained by microscopically visualized oil displacement experiments. Furthermore, the macroscopic interfacial properties, such as oil-water interfacial tension, interfacial rheological properties, coalescence of oil droplets of oil droplets, and viscosity of emulsions, were explored. The dynamic oil displacement images were utilized to investigate the characteristics of mobilization and migration of crude oil and the corresponding microscopic oil displacement mechanisms of betaine solutions with different hydrophobic branched chains. The following conclusions are drawn:

1) The adsorption of betaine molecules at the oil-water interface can not only significantly reduce the interfacial tension but also form a strong interfacial film with the emulsification and viscosity enhancement effects. The property of interfacial film can be regulated by the change in hydrophobic branched chains. As the branched degree of betaines increases, the strength of the interfacial film and the coalescence time decline and the emulsification and viscosity enhancement effects weaken.
2) In the homogeneous model, the strength of the interfacial film formed by betaines and crude oil increases, while the deformation ability of the oil droplets is poor. The oil recovery and sweep efficiency are improved by the in-situ plugging effect in the pore throat, and the remaining oil is mainly in the form of droplets. As the branched degree increases, the strength of the interfacial film declines, the increase in sweep efficiency slows down, and the oil recovery dwindles. The LSB solution with the highest sweep efficiency exhibits a superior oil displacement effect.
3) In the heterogeneous model, the plugging effect of oil droplets enhances the "local heterogeneity". Although the three betaines can improve the sweep efficiency, the displacement solutions still migrate along the dominant pathway after breaking through crude oil within the sweep range. A large amount of isolated cluster residual oil remains and this results in slightly lower oil displacement efficiency than that of water flooding. Therefore, oil recovery by betaines is close to that by water flooding in the heterogeneous model.

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Conflict of interest

The authors declare no competing financial interest.

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