

# Displacement Characters of Combination Flooding Systems consisting of Gemini-Nonionic Mixed Surfactant and Hydrophobically Associating Polyacrylamide for Bohai Offshore Oilfield

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**ABSTRACT:** In this paper, the injectivity and displacement characters of a new polymer–surfactant (SP) flooding system consisting of gemini–nonionic mixed surfactant and hydrophobically associating polyacrylamide (HAPAM) have been studied under Bohai offshore oilfield reservoir conditions. The injectivity tests have shown that the SP system could build higher resistance factor (RF) and residual resistance factor (RRF) to expand sweep efficiency in nonheterogeneous reservoirs. A total of 10 core flooding tests were carried out to investigate the effect of heterogeneity, slug composition, and polymer molecular structure. The results have shown that the system could obtain higher recovery owing to higher viscosity and ultralow interfacial tension (IFT) compare to water flooding and polymer flooding. The slug composition, with 0.175% polymer and 0.3% mixed surfactant, which forms a composite with the ratio of 4:1 gemini surfactant to nonionic surfactant, could allow full play to viscosity and IFT economically. Moreover, we found that the SP system with HAPAM could obtain a higher oil recovery by 6–13% original oil in place (OOIP) than a nonmodified one, as a result of viscoelastic rheology. The laboratory core tests provided credible proofs for large scale application with this SP system in Bohai offshore oilfield.

## 1. INTRODUCTION

At present, newly discovered oil fields have been declining steadily, so a considerable portion of oil production comes from mature fields. Generally, these oilfields have higher water cut after several years of exploring. Therefore, the chemical flooding with polymer could improve their recovery, owing to reducing the mobility ratio between the water and the oil, such as polymer flooding,<sup>1–4</sup> alkali–polymer–surfactant (ASP),<sup>5–8</sup> and polymer–surfactant (SP) flooding.<sup>9,10</sup> In addition, the main focus has been brought to other difficult exploring reservoirs in the past, such as complicated geologic reservoirs, offshore oil fields, heavy oil pools, and higher temperature and salinity reservoirs.<sup>4,11,12</sup>

Bohai offshore oilfield is the largest of all offshore oilfields in China; the oil recovery by water flooding is only 18–20% original oil in place (OOIP) compared to the average oil recovery of 32% OOIP in an onshore oilfield. The major reasons lie in the high oil viscosity (50–150 mPa s, average viscosity 70 mPa s), thick pay zones with high heterogeneity, high reservoir temperature (about 65 °C), high degree of mineralization (total dissolved substance (TDS), 9780–10890 mg L<sup>-1</sup>), and high hardness (Ca<sup>2+</sup>, 334–517 mg L<sup>-1</sup>; Mg<sup>2+</sup>, 145–228 mg L<sup>-1</sup>). Conventional polymers for improved oil recovery, such as partially hydrolyzed polyacrylamide (HPAM), cannot satisfy the exploring requirements. To solve the mentioned problems, polymer flooding with HAPAM has been carried out in SZ36-1 oilfield from 2003, and it has shown significant effect on incremental oil and decremental water cut.<sup>10,13</sup>

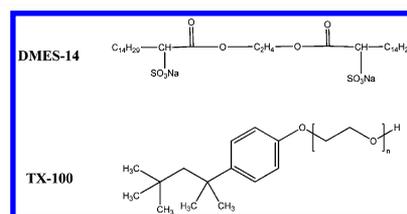


Figure 1. Structures of DMES-14 and TX-100.

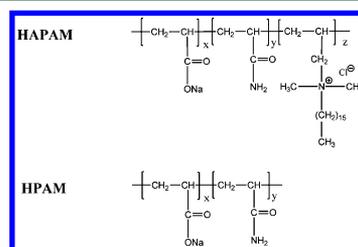


Figure 2. Structures of HAPAM and HPAM.

However, the recovery efficiency with polymer flooding still cannot satisfy the requirement of large scale exploitation in Bohai offshore oilfield. To reduce economical and technical risks of developing offshore oilfields, higher exploitative speed and higher recovery efficiency are the primary targets.<sup>12</sup>

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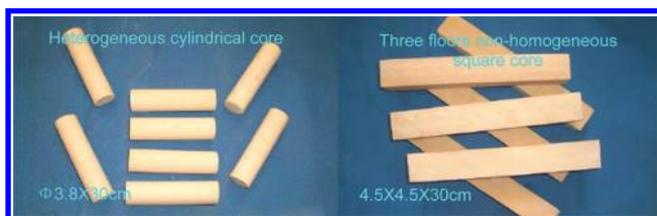
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Combination flooding systems, such as ASP and SP flooding, are feasible methods and have been successfully applied in some onshore oilfields.<sup>5–9</sup> In our previous study,<sup>14</sup> the disadvantages of ASP flooding have been discussed. In addition,

**Table 1. Composition of the Injected Water**

ion style	ion concentration (mg L <sup>-1</sup> )
K <sup>+</sup> and Na <sup>+</sup>	3085.61
Ca <sup>2+</sup>	275.55
Mg <sup>2+</sup>	154.5
CO <sub>3</sub> <sup>2-</sup>	14.18
HCO <sub>3</sub> <sup>-</sup>	311.52
SO <sub>4</sub> <sup>2-</sup>	85.25
Cl <sup>-</sup>	5439.06
total dissolved substance (TDS)	9367.66



**Figure 3.** The aspect of artificial sand cores.

**Table 2. Cores Properties**

core no. <sup>a</sup>	D (cm)	L (cm)	PV (mL)	Φ (%)	K <sub>w</sub> (× 10 <sup>3</sup> μm <sup>2</sup> )	S <sub>0</sub> (%)
1	3.8	30	114.5	33.65	264.5	0
2	3.8	30	123.0	36.15	1049.7	0
3	3.8	30	118.0	34.68	1940.0	0
4	3.8	30	112.0	31.9	760.0	83.0
5	3.8	30	107.0	31.45	465.1	86.9
6	3.8	30	107.5	31.6	637.0	81.7
7	4.5	30	135.5	22.3	725.4 <sup>b</sup>	79.0
8	4.5	30	129.5	21.3	466.2 <sup>b</sup>	75.0
9	4.5	30	132.5	31.6	784.0 <sup>b</sup>	81.7
10	4.5	30	132.5	21.81	662.9 <sup>b</sup>	76.2
11	4.5	30	131.2	21.56	702.2 <sup>b</sup>	75.6
12	4.5	30	138.5	22.71	619.9 <sup>b</sup>	79.0
13	4.5	30	140.1	23.05	728.1 <sup>b</sup>	75.4

<sup>a</sup>1–6: heterogeneous cylindrical cores. 7–13: three floors non-homogeneous square cores (K<sub>w</sub> = 0.6 μm<sup>2</sup>, 2 μm<sup>2</sup>, 3.2 μm<sup>2</sup>).  
<sup>b</sup>Composite water permeability.

a SP flooding system consisting of gemini–nonionic mixed surfactant and HAPAM has been designed and evaluated. The displacing fluid, which has ultralow oil–water interfacial tension and higher viscosity, has also shown good adaptability to the reservoir conditions in Bohai offshore oilfield.

In this paper, a total of 13 core flood tests were carried out to investigate the displacement characters of this SP flooding system, including the injectivity, RF/RRF, and the effect of displacement efficiency on heterogeneity, slug composition, and polymer molecular structure.

## 2. EXPERIMENTAL SECTION

**2.1. Materials.** Anionic gemini surfactant DMES-14 was synthesized according to a patent;<sup>15</sup> nonionic surfactant TX-100 was purchased from Chengdu Kelong Chemical reagents corporation (China). Their molecular structures are illustrated in Figure 1.

HPAM was purchased from Japan, trade name MO4000, [average molecular mass, 25 000 KDa; hydrolysis degree, 17.6% (w/w)]. HAPAM is similar to HPAM except that it contains a very small portion of hydrophobic monomer, [intrinsic viscosity, 2624.9 mL g<sup>-1</sup>; hydrolysis degree, 23% (w/w), hydrophobic monomer content, 0.5% (mol/mol)]. It was synthesized according to the literature.<sup>16</sup> Their molecular structures were illustrated in Figure 2.

Other reagents were purchased from Chengdu Kelong Chemical reagents corporation (China), and all of reagents were used as received without further purification.

The crude oil was obtained from Bohai offshore oilfield SZ36-1-A platform, after dehydration, with density of 0.95 g/cm<sup>3</sup>. In core flooding tests, its viscosity was adjusted to 100 mPa s and 50 mPa s at reservoir temperature (65 °C) with added kerosene.

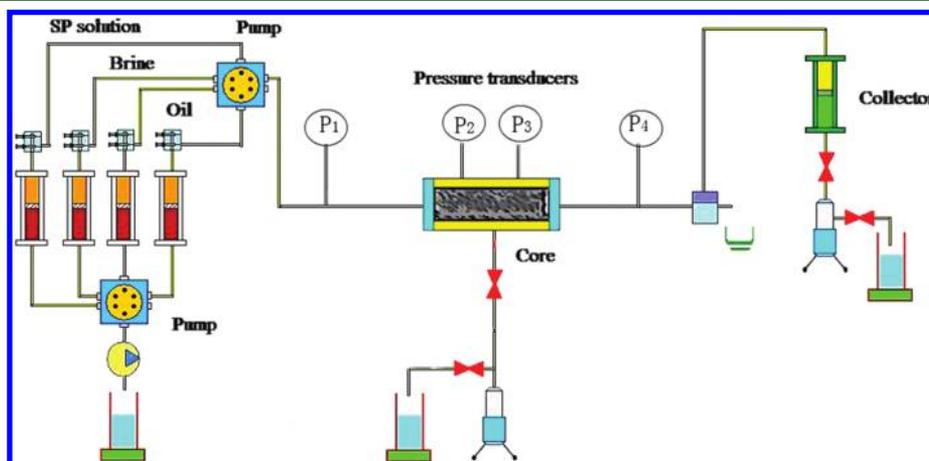
The brine was obtained from Bohai offshore oilfield SZ36-1-CEP platform and filtered with a 0.3 μm membrane of cellulose acetate. It was used for solution preparation and core tests. Its composition and salinity are given in Table 1.

All cores are artificial sand cores (Figure 3), and their properties are given in Table 2.

**2.2. Methods.** **2.2.1. Solution Preparation.** The surfactant or polymer stock solution of 5000 mg L<sup>-1</sup> was prepared by dissolving surfactants or polymer in brine. The dilute surfactants–polymer (SP) solution was prepared by mixing the stock solution in brine to obtain the desired surfactant and polymer concentrations.

**2.2.2. Measurement of Oil–Water Interfacial Tension.** The oil–water IFTs between solution and crude oil were measured using Texas-500C spinning drop tensiometer (Bowling, U.S.A.) for 30 min at 65 °C. The instrument could automatically record the interfacial tension with an image pick-up device and image acquisition software.

**2.2.3. Measurement of Viscosity.** The viscosities of the solutions were measured using Brookfield DV-III viscometer (Brookfield, U.S.A.) with a shear rate of 7.34 s<sup>-1</sup> at 65 °C.



**Figure 4.** Diagram of chemical flood system.

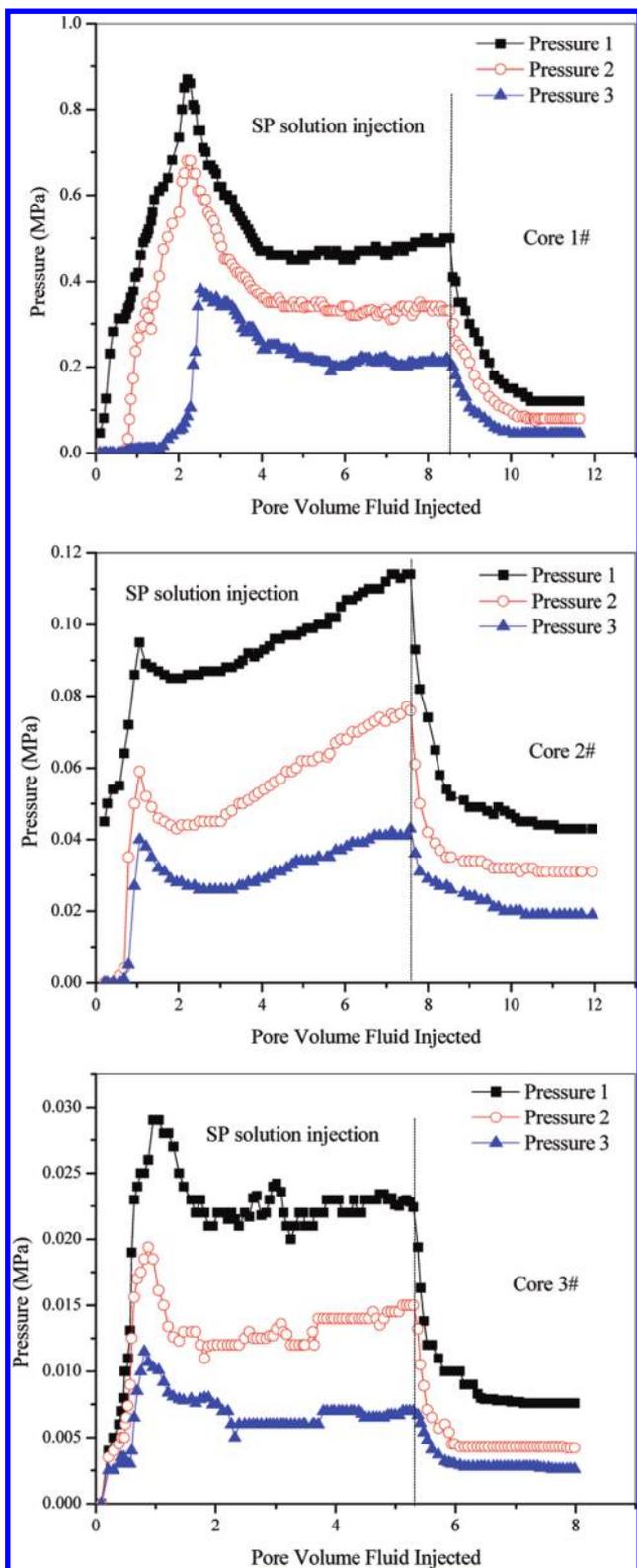


Figure 5. Relation of injective SP solution and pressure with different permeability.

2.2.4. *Preparation of Core Test.* The cores were dried at 70 °C for 24 h; their weights and sizes were measured, and then they were evacuated with a vacuum pump to less than 1 mmHg and saturated with brine to measure pore volume. Finally, pumping into brine at a rate of 1 mL/min, water permeability and porosity were calculated on basis of Darcy’s law.

Table 3. Summary of Core Flooding Tests

core no.	$K_w (\times 10^3 \mu\text{m}^2)$	RF	RRF
1	264.5	90.55	11.92
2	1049.7	63.55	23.97
3	1940.0	24.06	8.16

2.2.5. *Injectivity Test.* The cores saturated with brine were inserted into the core holder with length of 65 cm, diameter of 12 cm, and three pressure taps distributed along the core holder at a distance of 10 cm from the inlet and outlet of it. The displacing solutions were injected into the cores at constant rate of 1.0 mL/min until every pressure was steady. Subsequently, brine was injected into the cores, which had absorbed polymer and surfactant, and when the pressure drop stabilized across the cores, the experiment was finished. All the tests were run at 65 °C, and the pressures were recorded by a data terminal.

2.2.6. *Core Flooding Test.* Crude oil was injected continuously with a positive-displacement pump and an air bath held at 65 °C, until no more water was produced, then the water flooding was started until initial oil saturation was reached.

In Bohai offshore oilfield, considering economic reasons, the flooding method shifts from water flooding to chemical flooding when water cut reaches 75%. So, water flooding was first carried out until the water cut of the produced fluid reached 75%; then, the chemical flooding was carried out with injected 0.3 PV displacement plug, followed subsequently by chase water flooding to the water cut of produced fluid approaching 98%. The oil recoveries and water cuts were calculated every 0.15 PV during displacement. The flooding system is presented in Figure 4.

### 3. RESULTS AND DISCUSSION

3.1. *Injectivity.* Increased injected fluid viscosity could substantially reduce injectivity, consequently slowing fluid throughput and delaying oil production from reservoirs. Especially in lower permeability reservoirs, higher injected pressure may lead to the chemical flooding failure. Moreover, uniform pressure gradients imply the displacement fluid can effectively sweep core to increase mobility control capability.

Figure 5 presents the SP solution injectivity with various permeability heterogeneous cores. The injective pressures rose with PV of SP solution, then decreased to a steady realm where the three pressures were reproducible within 0.001 MPa. Moreover, the gradients of pressure drop are uniform and showed that the SP solution does not block the flow channel. With the decreasing core permeability, the highest of injected pressures improved from 0.03 to 0.9 MPa, and the peak delayed from 1.0 PV to 2.0 PV. This showed that this SP displacement fluid can well adapt to different permeability reservoirs.

3.2. *RF and RRF.* The resistance factor (RF) is the ratio between the differential pressure during chemical flooding and water flooding. It could be used to quantify the mobility reduction effect by increasing water viscosity and decreasing porous permeability. The residual resistance factor (RRF) is defined as the degree of permeability reduction of porous after chemical flooding. It is the ratio between the differential pressure of after and before chemical flooding. The RF and RRF could be calculated with eq 1 and eq 2:

$$RF = \frac{(\Delta P)_{SP}}{(\Delta P)_{wb}} \tag{1}$$

$$RRF = \frac{(\Delta P)_{wa}}{(\Delta P)_{wb}} \tag{2}$$

Using the data of the injective test, RF and RRF were calculated and presented in Table 3. The figures and table data

Table 4. Effect of Slug Composition and Fluid Properties on Oil Recovery

core no.	slug composition			properties of fluid before injecting			oil recovery (% OOIP)	
	$C_p$ (mg L <sup>-1</sup> )	$C_s$ (mg L <sup>-1</sup> )	proportion of DMES and TX-100	shearing time (s)	$\mu_w$ (mPa s)	IFT ( $\times 10^3$ m Nm <sup>-1</sup> )	water flooding	SP and sequent water flooding
4	1750	0	0	10	36.4		43.87	33.33
5	1750	3000	4:1	10	26.3	4.63	42.58	42.47
6	1750	3000	5:1	10	23.4	0.632	43.51	46.22
7	1750	0	0	10	36.4		18.88	27.29
8	1750	3000	4:1	10	26.3	4.63	14.32	38.60
9	1750	3000	5:1	10	23.4	0.632	16.72	35.02
10	1300	3000	4:1	0	21.6	3.36	27.13	35.25
11	1750	3000	4:1	0	51.0	4.17	22.42	36.26
12 <sup>a</sup>	1750	3000	4:1	0	23.0	3.14	22.84	22.39
13 <sup>a</sup>	2750	3000	4:1	0	47.5	3.72	23.98	30.14

<sup>a</sup>Polymer is HPAM (MO4000).

showed that the displacement system could quickly build flow resistance in higher permeability core during chemical flooding process. The characters illustrated that it could easily sweep to lower permeability floor to improve displace efficiency. This result is in agreement with the literature.<sup>17,18</sup> In addition, owing to the adsorption and retention of HAPAM on sand surface,<sup>19,20</sup> the SP system has higher RRF, decreasing permeability of higher permeability floor. These characters have shown that the SP system can improve oil recovery together with chemical flooding section with sequent water flooding section. It has been proven in a pilot test<sup>21</sup> and subsequent displacement experiments. We consider that the HAPAM solution structure, which was destroyed in the injecting process, could restore a supermolecular network in inaccessible porous volume (IPV) and other lower shear rate region, due to associative effects different from HPAM at lower polymer concentrations, as it was proven in the literature.<sup>22,23</sup>

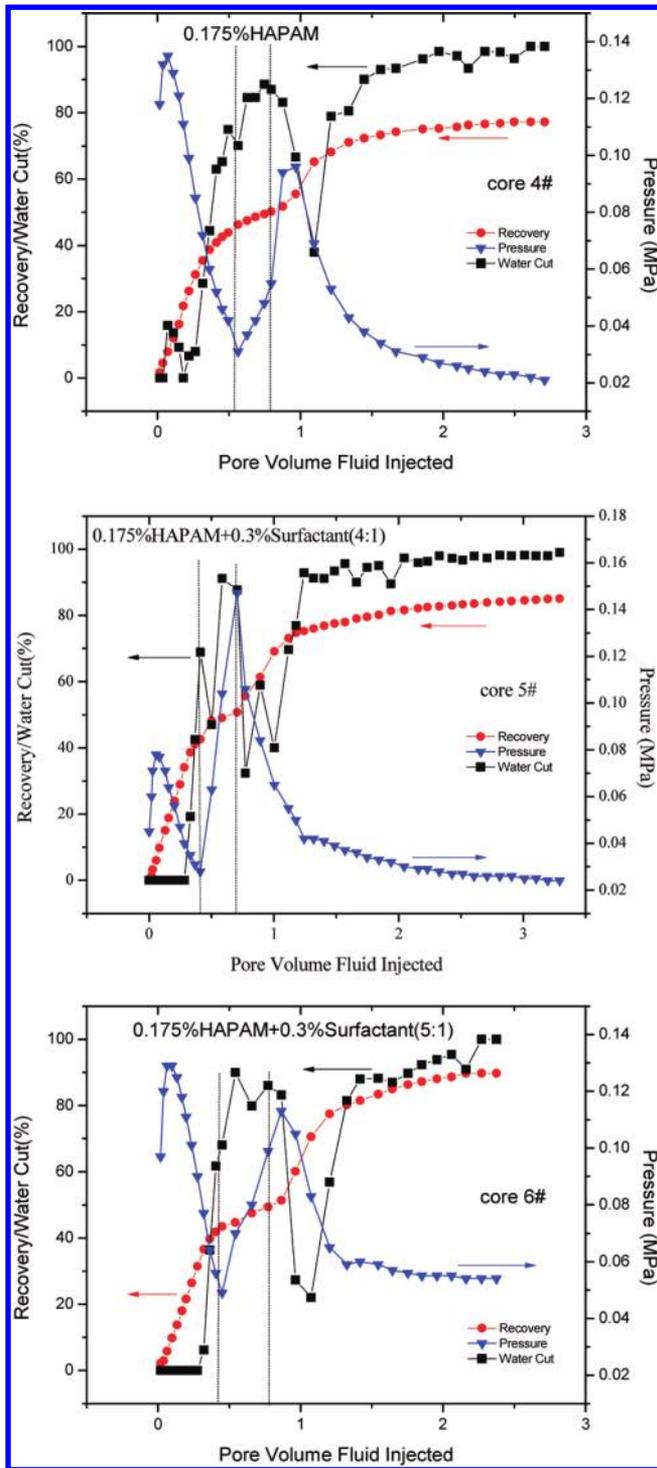
**3.3. Effect of Permeability Heterogeneity to Oil Recovery.** We used homogeneous cylindrical cores and three floors nonhomogeneous square cores ( $K_g = 0.6 \mu\text{m}^2$ ,  $2 \mu\text{m}^2$ ,  $3.2 \mu\text{m}^2$ ) with equal water permeability to investigate the effect of permeability heterogeneity on oil recovery. The permeability heterogeneity is defined with  $K_{\text{max}}/K_{\text{min}}$ . In these experiments, all values of  $K_{\text{max}}/K_{\text{min}}$  are equal to 5.3. Contrasting cores 4 and 7, 5 and 8, and 6 and 9, they have approximately equal water permeability, but due to the effect of heterogeneity, the homogeneous cores could obtain higher (about 35%) water flooding recovery than nonhomogeneous cores. However, this difference decreased to 6% in chemical flooding and sequent water flooding (Table 4). Because the displace system with HAPAM could obtain higher RRF than HPAM,<sup>24</sup> in the sequent water flooding section, it still modifies the water cut profile to promote more displaced fluid to enter into the part with lower permeability. The results illustrated that the polymer or SP flooding system with HAPAM could improve obviously oil recovery for nonhomogeneous reservoirs.

**3.4. Effect of Slug Composition to Oil Recovery.** The slug composition is a key factor to investigate the economy of chemical flooding system. In this section, the displaced fluid was sheared for 10 s before it was injected into the core to simulate the shearing process while the solution was injected into floor, the method was described in a previous study.<sup>14</sup> Contrasting the oil recovery of homogeneous cores (4, 5, and 6) and nonhomogeneous cores (7, 8, and 9) (Table 4), we discovered common rules.

Figure 6 shows the change in oil recovery with surfactant composition at a fixed polymer concentration of 0.175%. The core 4 test used polymer flooding without surfactant, which could improve recovery of 33.33% with chemical flooding and sequent water flooding. The other slug compositions 5 and 6 contain 0.3% mixed surfactant with a ratio of DMES-14 and TX-100 equal to 4:1 and 5:1, respectively. In a previous study, we have already discovered that the IFTs decreased with the increased proportion of DMES-14.<sup>14</sup> In these tests, their oil recovery can reach up to 42.47% and 46.22%. Although the SP system viscosity decreased from 36.4 mPa s to 23.4 mPa s when surfactants were appended, the mixed surfactant still contributed with an approximate oil recovery of 10%. Contrasting cores 5 and 6, despite the fact that a higher ratio DMES-14 to TX-100 could decrease IFT to  $10^{-4}$  mN/m, the oil recovery increased only 2% compared to the case of the lower ratio. The recovery of nonhomogeneous core tests showed similar rules (Figure 7), but the use of 4:1 DMES-14/TX-100 reached higher recovery than 5:1 system, because the former has better mobility control capability in nonhomogeneous core with higher viscosity. These core tests illustrated that slug composition with 0.175% polymer and 0.3% mixed surfactant with a ratio of 4:1 DMES-14 to TX-100 could reach higher oil recovery. Furthermore, appropriate composition of the SP system could be matched to permeability heterogeneity reservoirs.

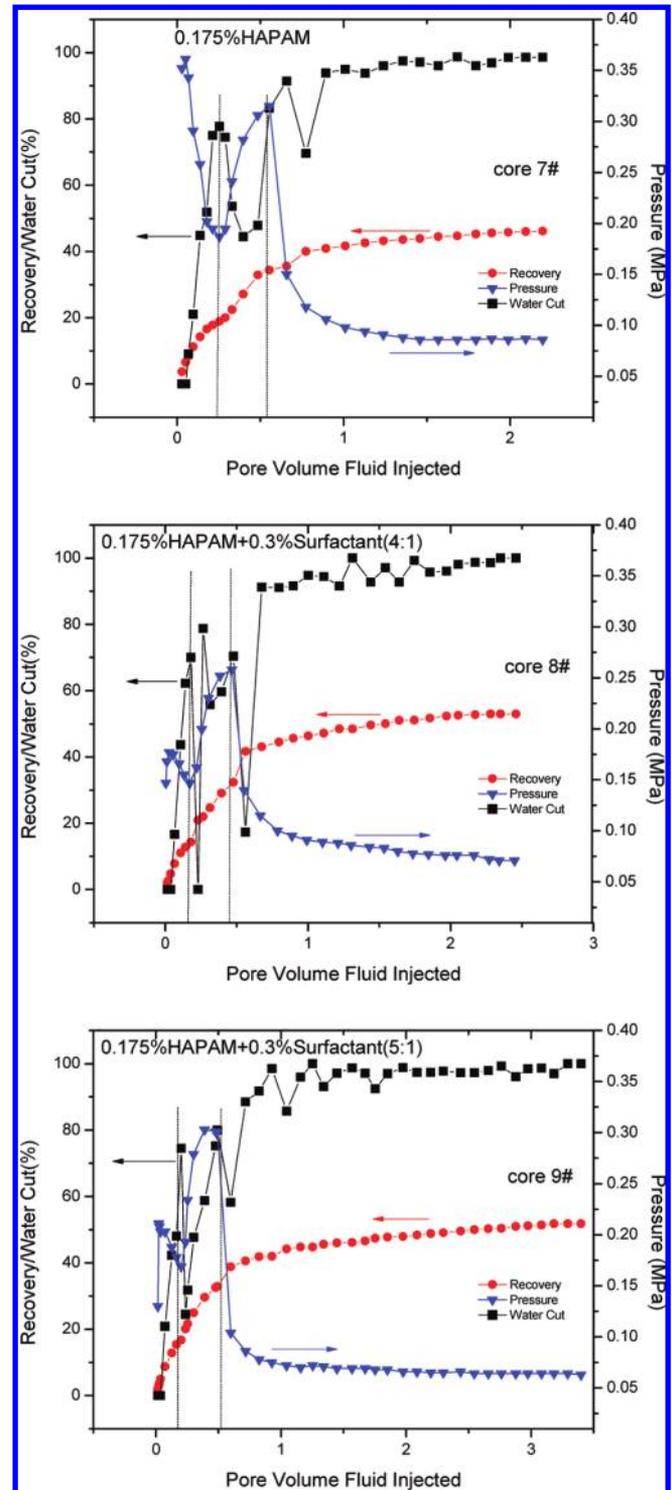
**3.5. Effect of Polymer Structure of SP System to Oil Recovery.** Though in many articles in the literature<sup>23,24</sup> the different displacement characters between HPAM and HAPAM had been discussed, their differences were seldom studied when applied in SP or ASP flooding. So, we designed two kinds of SP flooding systems with different molecular structure polymer (HPAM and HAPAM) to investigate the effect of polymer structure on oil recovery (Figure 8 and Table 4).

Cores 10 and 11 were submitted to injection of HAPAM and mixed surfactant (4:1 DMES-14/TX-100), while in the cores 12 and 13 were used HPAM and the same surfactants composition as in cores 10 and 11. The viscosity of crude oil was 50 mPa s at 65 °C with added kerosene. When the two SP systems composed with uniform concentration of polymer and surfactants were used (cores 11 and 12), the SP system with HAPAM increased oil recovery to 36.26% during SP flooding and sequent water flooding. However, the SP system with HPAM only increased recovery to 22.39% because the former could offer higher viscosity to modify water cut profile. Contrasting the different SP systems that have common viscosity and IFT (10 and 12; 11 and 13), we still find that



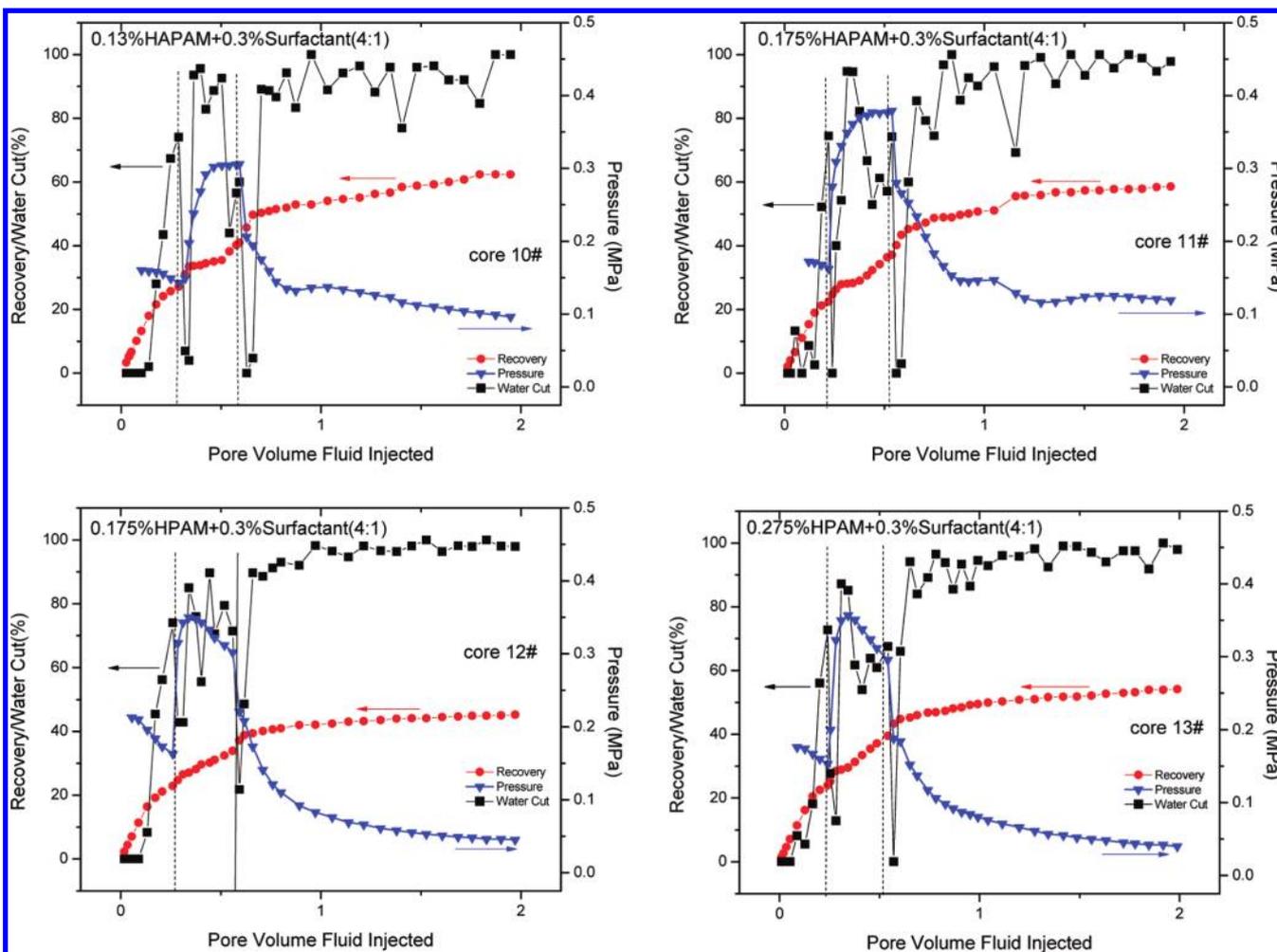
**Figure 6.** Cumulative oil recovery, water cut, and pressure history of different slug composition (homogeneous permeability cores): 0.3 PV, 65 °C,  $\mu_o = 100$  mPa s.

the SP system with HAPAM could obtain higher oil recovery by 6% to 13% OOIP. Moreover, when the viscosity of the SP system increases from 23 to 50 mPa s (10 and 11; 12 and 13), the SP system with HPAM could increase oil recovery by 7.7% and the SP system with HAPAM could improve only 1%. This is an interesting phenomenon, because it means that the SP system with HAPAM could increase oil recovery with other mechanisms different from HPAM.



**Figure 7.** Cumulative oil recovery, water cut, and pressure history of different slug composition (nonhomogeneous permeability cores): 0.3 PV, 65 °C,  $\mu_o = 100$  mPa s.

Generally, the combination flooding systems like ASP or SP increase oil recovery with extent sweep efficiency by higher viscosity and increasing wash efficiency by lower IFT. It is known that the associating polymer could obtain higher viscosity because intermolecular association would lead to a supermolecular network, in the semidilute regime, where the solution is viscoelastic. Moreover, in different flow filed, viscoelastic fluid can show elastic deformation. When the



**Figure 8.** Cumulative oil recovery, water cut, and pressure history of SP flooding with different polymer (nonhomogeneous permeability cores): 0.3 PV, 65 °C,  $\mu_o = 50$  mPa s.

associating polymer solution flow in channels of porous media with different diameter, the deformation of fluid could “drag” oil drop from holes in core to improve oil recovery, the mechanism already was partly confirmed with our microcosmic flooding experiments.

#### 4. CONCLUSIONS

The SP solution that was composed with HAPAM, gemini surfactant DMES-14, and nonionic surfactant TX-100 has shown excellent viscosity and IFT properties under the condition of oil reservoir. This SP flooding system could form higher resistance factor and residual resistance factor to modify water cut profile, especially in nonhomogeneous reservoirs.

By the injection of the 0.3 PV displacement solution, the polymer flooding recovery could improve 30% (OOIP) more than water flooding and the SP flooding recovery could improve about 10% (OOIP) more than the polymer flooding.

Through a series of core flooding tests, the SP flooding could decrease effectively oil–water interfacial tension to increase obviously oil recovery comparing with water flooding and polymer flooding.

Contrasting the SP flooding with HAPAM and HPAM, the former can obtain higher oil recovery about 6% to 13% OOIP than the latter. We consider that the HAPAM could form

supermolecular network to strengthen viscoelastic rheological properties to increase oil recovery.

The core experiments proved that the SP displacement system is suitable for the Bohai offshore oilfield explorer with chemical flooding.

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##### Notes

The authors declare no competing financial interest.

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#### ■ NOMENCLATURE

HAPAM = hydrophobically associating polyacrylamide  
 HPAM = partially hydrolyzed polyacrylamide  
 RF = resistance factor  
 RRF = residual resistance factor  
 IFT = interfacial tension  
 TDS = total dissolved solid

$D$  = diameter of core, cm

$L$  = length of core, cm

PV = pore volume, cm<sup>3</sup>

$\Phi$  = porosity of core, percent

$K_w$  = water permeability of core,  $\mu\text{m}^2$

$K_g$  = gas permeability of core,  $\mu\text{m}^2$

$S_0$  = initial oil saturation, percent

$C_p$  = polymer concentration, mg L<sup>-1</sup>

$C_s$  = total surfactant concentration, mg L<sup>-1</sup>

$\mu_w$  = viscosity of aqueous fluid, mPa s

$\mu_o$  = viscosity of oil, mPa s

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