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Oil Recovery at High Brine Salinity Conditions Using Amphoteric Terpolymer

The viscosity and the oil recovery ability of high molecular weight amphoteric terpolymer (ATP) were tested in different brines and compared with hydrolyzed poly(acrylamide) (HPAM) which is traditionally used in enhanced oil recovery (EOR). The results show that ATP provides higher viscosity at brine salinity ranging from 200 to 300 g·L^{-1}. This may give ATP an advantage over HPAM in high salinity reservoirs, which are abundant in Kazakhstan. Moreover, comparative polymer flooding experiments were carried out by injecting 0.25 wt.% ATP and HPAM dissolved in 200 g·L^{-1} brine into sand packs. The HPAM flooding in high permeability (1.77 Darcy) sand pack leads to the increase of the oil recovery factor (ORF) by 18.5 % after 1 PV of brine injection. On the other hand, the injection of ATP into the low permeability (0.6 Darcy) sand pack results in the ORF increase by 28.5 %. The obtained results show that ATP has a potential to become an alternative to HPAM in 200 g·L^{-1} and higher brine salinity reservoirs.

Keywords: amphoteric terpolymer, hydrolyzed poly(acrylamide), antipolyelectrolyte effect, sand pack model, water flooding, polymer flooding, oil recovery factor.

Introduction

Water is usually used to displace oil from matrix rocks. However, because of the unstable displacement front due to the differences in oil and water viscosities and heterogeneous nature of matrix rocks the oil production rates are often decline accompanied by the increase of water production [1].

If water viscosity is increased by dissolving high molecular weight polymers in it, the problem of viscous fingering and premature breakthrough can be minimized [2]. Such polymers are an important topic of modern research due to their practical application. For example, polyacrylamide is widely used in wastewater treatment, paper production and oil industry because of its thickening, flocculation and rheological properties [3, 4].

Even though different polymers were tested over the years, hydrolyzed polyacrylamide (HPAM) has found the widest application due to its low cost; commercial availability; remarkable capability to increase viscosity; acceptable and adjustable injectivity; and resistance to microbial degradation [5].

However, if cations are present in water the negative charges on the polymer chain are screened and as a result the hydrodynamic volume of the polymer molecule is reduced [5, 6]. Thus, at higher salinities higher concentrations of HPAM are required in order to achieve the target viscosity. Moreover, at extremely high salinity and temperature the HPAM chains will coil up and precipitate [6].

Amphoteric copolymers and terpolymers can swell and enhance the viscosity in high salinity reservoirs, which is crucial for enhanced oil recovery (EOR) [7, 8]. The remarkable resistance of polyampholytes to high salinity makes them attractive for the application where thickeners are required in concentrated brine solutions [9-14]. A special attention should be given to the hydrophobically associating polyampholytes which combine self-assembly and adjustable charge balance [15–18]. The application of acrylamide-based polyampholytes in EOR and drag reduction was reviewed in [19].

In this regard, amphoteric polyelectrolytes have great potential, because in high-salinity water anions and cations disperse macromolecular chains by reducing the electrostatic attraction between positively and negatively charged monomers. As a result, the viscosity of the solution increases [20]. The main problems of oil fields in Kazakhstan are the viscosity of oil and high salinity of reservoir water, which in some regions reaches 163–232 g·L^{-1} and even higher. To solve these problems, it is recommended to use specially designed amphoteric therpolymers (with "anti-electrolyte" effect) that can increase the viscosity of brine [21–26].

In this study we compare the oil recovery efficiency of high molecular weight amphoteric terpolymer AAm-AMPS-APTAC = 80:10:10 mol.% with HPAM in sand pack models at high salinity conditions.

Experimental

Materials

Monomers – acrylamide (AAm, 97 % purity), 2-acrylamido-2-methylpropanesulfonic acid sodium salt (AMPS, 50 wt.%), (3-acrylamidopropyl) trimethylammonium chloride (APTAC, 75 wt.% in water), and ammonium persulfate (APS, 98% purity) were purchased from Sigma-Aldrich Chemical Co., and used without further purification. Commercially available hydrolyzed polyacrylamide (HPAM) (Flopaam 3630S, 98% purity SNF) with hydrolysis degree of 30% and average molecular weight of 17.2 million Dalton was used as received.

Sand pack model

To simulate the oil displacement by water and polymers, 3-cm-diameter and 5-cm-length sand packs were used in this study. The size of the sand grains varied in between 0.25 and 0.5 mm. Initially the sand packs were vacuumed and saturated with brine. The porosity of the models was determined by using the following formula:

$$\varphi = \frac{V_{in} - V_d}{V_t} \cdot 100\,\%,$$

where φ — is the porosity, %; V_{in} — is the injected volume, cm^3; V_d — is the dead volume, cm^3; V_t — total volume of the sand pack, cm^3

As the next step, brine was injected though the sand packs at different flow rates and constant registration of pressure. Darcy equation was used to calculate permeability at 100 % water saturation. The results of the porosity and permeability calculations are presented by Table 1.

Table 1

Properties of the sand packs

Sand pack	Pore volume, cm^3	Porosity, %	Permeability, Darcy
1	10.6	30	0.62
2	11.9	33.7	1.77

Brine

Brines with different salinities and chemical composition are listed in Table 2.

Table 2

Total salinity and chemical composition of synthetic brines

Total salinity, g·L^{-1}	Concentration of salts, g·L^{-1}		
	NaCl	$CaCl_2$	$MgCl_2$
200	180	10	10
232	208.8	11.6	11.6
250	225	12.5	12.5
275	247.5	13.75	13.75
300	270	15	15

Oil

Crude oil from Karazhanbas oilfield well #1913 was used. Oil viscosity and density at 30°C are equal to 420 mPa·sec and 0.93 g·cm^{-3}, respectively.

Methods

Elemental analysis of ATP and HPAM samples were performed using Vario EL-III elemental analyzer (Elementary Analyze System GmbH, Hanau, Germany). The dynamic viscosity of polymer solutions was measured by Ubbelohde viscometer at 24 and 60 °C. The sand pack experiments were done with the help of core flooding apparatus “УИК-С (2)” (Russia).

Results and Discussion

Synthesis and characterization of AAm-APTAC -AMPS and HPAM

Synthesis and characterization of the AAm-AMPS-APTAC amphoteric terpolymer containing 80 mol.% AAm, 10 mol.% AMPS and 10 mol.% APTAC with the weight-average molecular weight ($M_w = 2.9 \cdot 10^6$ Dalton) and the average-number molecular weight ($M_n = 2.1 \cdot 10^6$ Dalton) are comprehensively described elsewhere [24, 25] (Fig. 2).

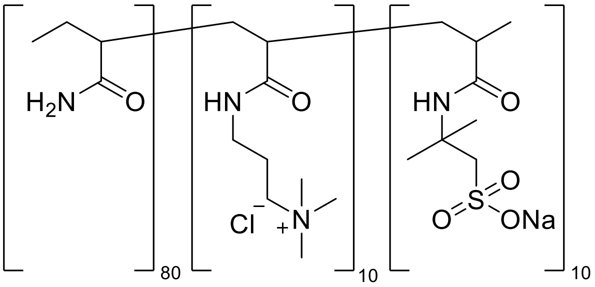
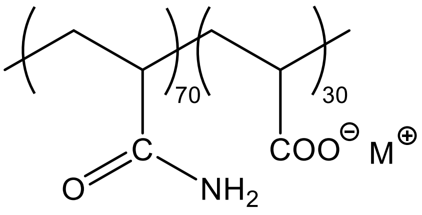


Figure 2. Repeating monomeric units and composition (mol.%)
of AAm-AMPS-APTAC amphoteric terpolymer

Commercially available hydrolyzed polyacrylamide (HPAM) (Flopaam 3630S, 98% purity, SNF) with a hydrolysis degree of 30 % and an average molecular weight of $17.2 \cdot 10^6$ Dalton was used for the comparative experiments. The hydrolysis degree of HPAM 30 % means that the sample contains 70 mol. % of acrylamide (AAm) and 30 mol.% of sodium acrylate (SA) in the macromolecular chain (Figure 3).



where M^+ is Na^+ or K^+

Figure 3. Repeating monomeric units and composition (mol.%) of HPAM

The results of the elemental analysis of AAm-AMPS-APTAC and HPAM are presented in Table 3.

Table 3

Results of elemental analysis of AAm-AMPS-APTAC and HPAM

Samples	Monomer ratios in terpolymer and copolymer, mol. %			Percentages of elements, wt. %				
				Found/Calculated	N, %	C, %	H, %	S, %
ATP	AAm	APTAC	AMPS	Found	11.54	43.58	8.32	3.18
	80	10	10	Calculated	15.34	47.83	7.11	3.19
HPAM	AAm	SA		Found	8.49	38.29	6.46	0
	70	30		Calculated	11.84	43.52	5.36	0

The Table 3 shows that the experimentally found and theoretically calculated amounts of elements C, N, H and S are in good agreement. This confirms that the composition of the AAm-APTAC-AMPS (80:10:10 mol.%) terpolymer insignificantly deviates from the initial monomer composition in the feed. In case of HPAM, the experimentally found and theoretically calculated amounts of elements C, N and H are somewhat different. This is reasonable because the commercially available samples for oil recovery usually contain some impurities.

Viscosity of AAm-APTAC-AMPS and HPAM in synthetic brine

Figure 5 demonstrates the dynamic viscosities of amphoteric terpolymer AAm-AMPS-APTAC and HPAM versus the polymer concentrations at 24 and 60 °C in 250 g·L^{-1} synthetic brine. The gradually decreasing dynamic viscosities of ATP and HPAM upon dilution testify the absence of polyelectrolyte effect leading to the unfolding of macromolecular coils.

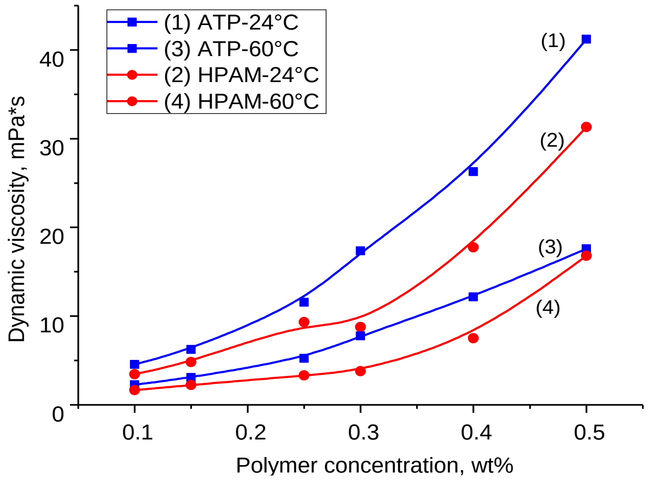


Figure 5. Dynamic viscosity of ATP and HPAM versus the polymer concentration in 250g·L^{-1} synthetic brine at 24 and 60 °C

Figure 6 represents the dynamic viscosities of ATP and HPAM in brine solutions. The dynamic viscosity of 0.25 wt.% ATP solutions increases steadily from 7.45 to 14.25 mPa·s with the increase in the salinity from 200 to 300 g·L^{-1}. Whereas the dynamic viscosity of HPAM solution increases from 6.78 to 9.31 mPa·s upon the salinity increase from 200 to 250 g·L^{-1}, however the further increase in the salinity up to 300 g·L^{-1} causes sharp falling of HPAM viscosity. This is explained by the precipitation of HPAM as a result of "salting out" effect.

The increase of the dynamic viscosity of amphoteric terpolymer in high saline solution is explained by the screening of the positively and negatively charged monomers by anions and cations of salts, which leads to the unfolding of macromolecular chains. This phenomenon is called *antipolylectrolyte effect*. The dynamic viscosities of ATP solutions are higher than that of HPAM. It is explained by the unfolding of AAm-AMPS-APTAC terpolymer molecule at high temperature and salinity.

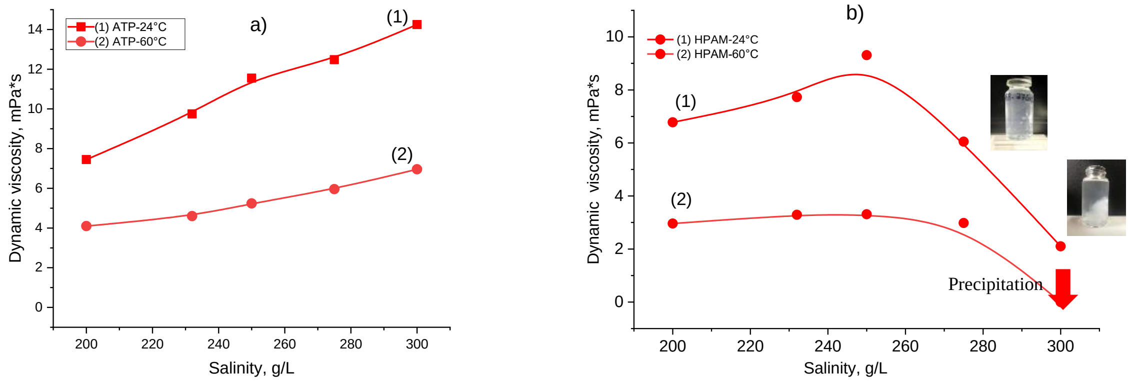
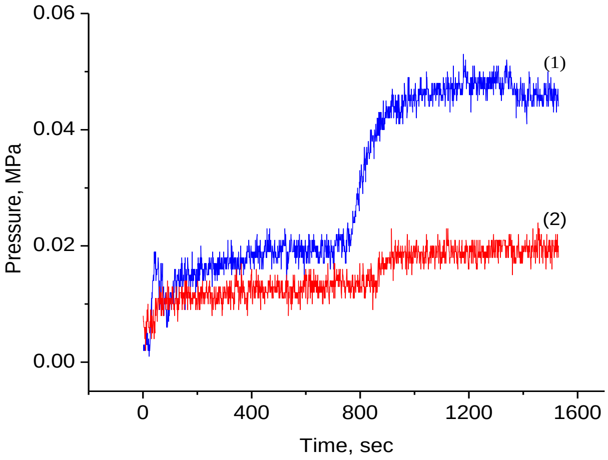


Figure 6 Salt-dependent dynamic viscosities of 0.25 wt.% amphoteric terpolymer (*a*) and HPAM (*b*) at 24°C (*1*) and 60°C (*2*) in the range of 200–300 g·L^{-1} brine salinity

Sand pack flooding

Water and oil saturation

The saturation of the models with 200 g·L^{-1} brine was done after vacuuming in order to calculate the porosity (Table 1). Figure 7 shows the injection pressure change during the oil saturation process for the 0.62 D and 1.77 D sand packs. As can be seen, for the low-permeability sand pack model, the oil injection pressure reached 0.044–0.051 MPa and stabilized without further growth. For the high-permeability sand pack model, the oil injection pressure stabilized at 0.020–0.024 MPa. This is two times less than the oil saturation pressure for the low-permeability sand pack model. In addition, the mass of saturated oil in each model was calculated on the basis of the material balance, and equal to 8.78 and 9.67 g for the 0.62D and 1.77D sand packs, respectively.

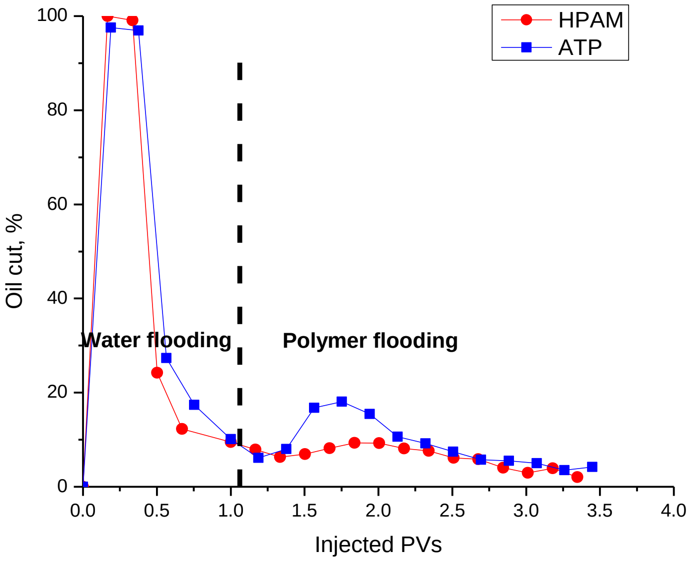


The permeability of sand pack model is 0.62D (*1*) and 1.77D (*2*). Flow rate — 0.15 cm^3·min^{-1}. Temperature — 30 °C

Figure 7. The oil injection pressure versus time during the oil saturation process

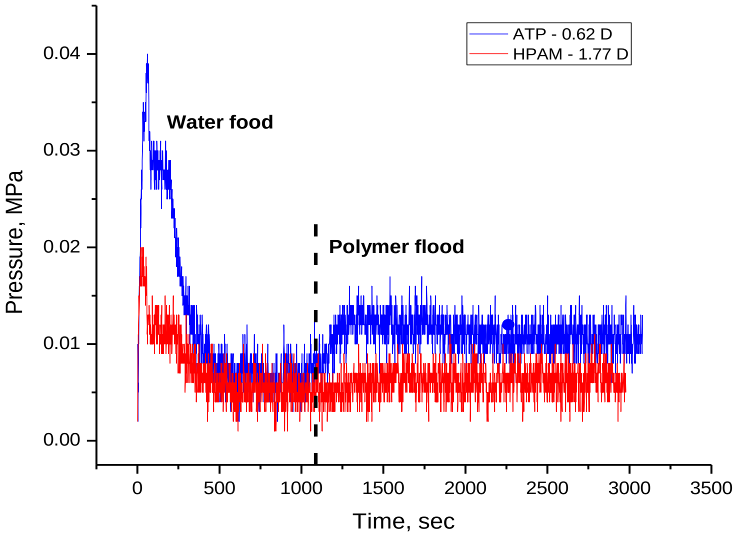
Water and polymer flooding

Figure 8 shows the oil cut versus the number of injected pore volumes. Water injection resulted in the production of almost the same portions of oil in both tests, even though in the case of low permeability (0.62D) sand pack, the injection pressure was significantly higher (Figure 9). The injection of the polymer solutions into this models shows that ATP provides a higher increase in the oil production than HPAM does.



Flow rate — 0.15 cm^3·min^{-1}. Temperature — 30 °C

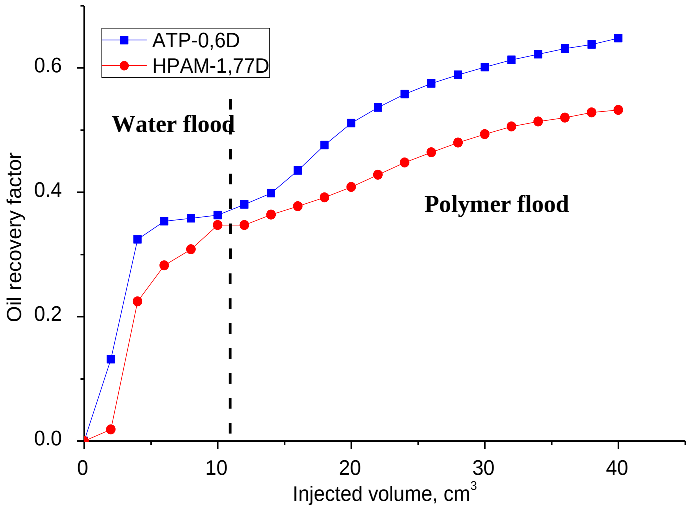
Figure 8. Oil cut versus injected pore volumes during water injection and polymer flooding by ATP (-■-) and HPAM (-●-)



The permeability of sand pack model is 0.62D (*1*) and 1.77D (*2*). Flow rate — 0.15 $cm^3 \cdot min^{-1}$. Temperature — 30 °C

Figure 9 The injection pressure versus time during water and polymer flooding

Figure 10 shows the oil recovery factor (ORF) obtained during water and polymer flooding processes. Water injection allowed to produce 34.7–36 % of oil. Whereas the injection of 0.25 wt.% ATP and HPAM solutions in 200 $g \cdot L^{-1}$ brine into the 0.6D and 1.77D sand packs increased the oil recovery factor after water injection by 28.5 % and 18.5 %, respectively. Thus, 0.25 wt.% ATP produced 10 % more oil than HPAM.



Flow rate — 0.15 $cm^3 \cdot min^{-1}$. Temperature — 30 °C

Figure 10. Oil recovery factor registered during ATP (- ■ -) and HPAM (–●–) polymer flooding experiments.

Conclusions

The results of the elemental analysis demonstrated that the composition of AAm-AMPS-APTAC terpolymer insignificantly deviates from the composition of initial monomer feed upon polymerization.

AAm-AMPS-APTAC terpolymers exhibited improved viscosifying behavior in high salinity media which is explained by their polyampholytic nature and can find practical applications in EOR. For example, the increase of brine salinity from 200 to 300 $g \cdot L^{-1}$ at 24 °C results in the increase of the dynamic viscosity of 0.25 % ATP solution from 7 to 14 mPa·s. Whereas, the dynamic viscosity of HPAM solution increases from 6 to 9 mPa·s upon the salinity increase from 200 to 250 $g \cdot L^{-1}$ and then suddenly falls down at 300 $g \cdot L^{-1}$ because of the severe polymer precipitation. This phenomenon is explained by hydrolyzed polyacrylamide poor solubility at 270–300 g·L^{-1}. The obtained viscosity measurement results clearly demonstrate that the amphoteric terpolymer is more stable at high brine salinity than HPAM.

The injection of 0.25% amphoteric terpolymer and HPAM solutions prepared in 200 g·L^{-1} brine into the 0.62 and 1.77 Darcy sand packs resulted in the increase of the oil recovery factor by 28 and 18%, respectively. These results show that the amphoteric terpolymer has a higher oil displacement capacity than HPAM.

The introduction of the amphoteric terpolymer as an alternative to HPAM at extremely high salinity conditions adds to the relevance and novelty of this work.

However, some significant shortcomings of this work are:

1) Limited number of sand pack flooding experiments;

2) The use of models with relatively low pore volume, which increase the probability of errors in oil recovery calculations;

3) No polymer retention measurements were done.

Thus, the future research will focus on the flooding tests with polymer retention measurements by using large pore volume sand packs and cores.

Acknowledgments

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Н. Мухаметгазы, И.Ш. Гуссенов, А.В. Шахворостов, Хейкки Тенху

Амфотерлік терполимерді қолдану арқылы жоғары тұзды орта жағдайында мұнай өндіру

Жоғары молекулалық салмақты амфотерлі терполимердің (АТП) тұтқырлығы мен мұнайды ығыстырып шығару қабілеті әртүрлі тұзды ерітінділерде сыналған және мұнайдың өнімділігін арттыруда дәстүрлі түрде қолданыстағы гидролизденген полиакриламид (ГПА) мен салыстырылған. Нәтижелер АТП 200-ден 300 г/л-ге дейінгі тұзды ерітінділерде жоғары тұтқырлықты қамтамасыз ететінін көрсетеді. Бұл Қазақстанда көптеп кезігетін тұздылығы жоғары су қоймаларында ГПА-мен салстрғанда АТП-ға елеулі артықшылық бере алады. Сонымен қатар, Полимерлік айдау бойынша салыстырмалы тәжірибелер 0.25 % массалық үлестегі амфотерлі терполимер мен ГПА-ның 200–300 г/л тұзды судағы ерітінділерін құмды модельге айдау арқылы жүргізілді. ГПА ерітіндісімен жоғары өткізгішті (1.77 Дарси) құмды модель арқылы айдау 1PV тұзды сумен айдауға салыстырғанда мұнай өндіру коэффициентін (МӨК) 18.5%-ға арттырды. Екінші жағынан, өткізгіштігі төмен (0.6 Дарси) құмды модельге АТП айдау МӨК-ті 28.5%-ға дейін артырды. Алынған нәтижелер АТП-ның 200 г/л және одан да жоғары тұзды су қоймаларында ГПА -ға балама бола алатынын көрсетеді.

Кілт сөздер: амфотерлік терполимер (АТП), гидролизденген полиакриламид, антиполиэлектролиттік эффект, құмды модель, су арқылы айдау, полимер ерітіндісімен айдау, мұнай өндіру коэффиценті.

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Добыча нефти в условиях высокой минерализации с применением амфотерного терполимера

В этой работе были протестированны реологические свойства и нефтевытесняющая способность высокомолекулярного амфотерного терполимера (АТП) в водах с различной минерализацией. Результаты сравнивались с частично гидролизованным полиакриламидом (ЧГПА), который традиционно используется для увеличения добычи нефти. Результаты показали, что АТП обладает большей вязкостью в диапазоне минерализации от 200 до 300 г/л. Это даёт АТП преимущество над ЧГПА в пластах с высокой минерализацией воды, коих в Казахстане довольно много. Сравнительные эксперименты по полимерному заводнению проводились путём закачки 0.25 мас.% амфотерного терполимера и ЧГПА, растворенных в воде с минерализацией 200 г/л в насыпные песчаные модели. Закачка ЧГПА в высокопроницаемую модель (1.77 Дарси) привела к увеличению коэффициента вытеснения нефти (КВН) на 18.5% после закачки одного порового объёма воды. Соответственно, при закачке амфотерного терполимера в низкопроницаемую (0.6 Дарси) модель КВН был увеличен на 28.5%. Полученные результаты показали, что АТП может стать альтернативой ЧГПА в условиях где минерализация воды превышает 200 г/л.

Ключевые слова: амфотерный терполимер (АТП), гидролизванный полиакриламид, антиполиэлектролитный эффект, насыпная модель, заводнение, полимерное заводнение, коэффициент вытеснения нефти.

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