

UDC 622.276.43:678.745.842

Article / Статья

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ANALYSING THE EFFICIENCY OF FLOODING OIL RESERVOIRS WITH WATER-SOLUBLE POLYACRYLAMIDE AND PRELIMINARY CROSS-LINKED POLYACRYLAMIDE PARTICLES

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АНАЛИЗ ЭФФЕКТИВНОСТИ ПРИМЕНЕНИЯ ЗАВОДНЕНИЯ НЕФТЕНОСНЫХ ПЛАСТОВ НА ОСНОВЕ ВОДОРАСТВОРИМОГО ПОЛИАКРИЛАМИДА И ПРЕДВАРИТЕЛЬНО СШИТЫХ ПОЛИАКРИЛАМИДНЫХ ЧАСТИЦ

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Received / Получена: 04.04.2019. Accepted / Принята: 01.08.2019. Published / Опубликовано: 27.09.2019

Keywords:

polymer flooding, polyacrylamide, absorbent, rheological characteristics, water inflow limitation, injectivity profile alignment

Due to the constant increase in the number of fields entering the final stage of operation, oil-producing companies are raising their demand for integrated technologies that can increase the coverage of the formation by the displacement process and reduce the water cut of well products at the same time. A traditional technology used for this purpose is flooding with viscous solutions of water-soluble polyacrylamide. The paper provides an overview of polymer flooding technologies, identifies the advantages and disadvantages of this approach, in particular, lists the factors that can influence the rheology of a polymer solution. Despite the extensive use of flooding based on polyacrylamide gels, this method has a number of limitations, primarily associated with the rheological characteristics of the polymer. The instability of such rheological characteristics of the polymer is a significant obstacle on the way to achieving the target oil recovery factor.

An alternative way of injectivity profile alignment is the flooding technology using polymer gel particles based on a suspension of cross-linked polyacrylamide particles. Such a polymer is synthesized in advance to form a three-dimensional structure (3D polymer). Backed by the literature review, the effectiveness of technologies based on water-soluble polyacrylamide and preliminary cross-linked polyacrylamide particles was compared; as a result, a conclusion was made about the superiority of the latter by a number of significant indicators. Polymers based on polymer gel particles do not change their rheological characteristics during the injection process, while the particles can change their shape and tear, enveloping the obstacles. Besides, the technology injecting the suspension with a smaller volume of reagent, successively selecting the optimal particle size distribution depending on the specific geotechnical conditions of the formation. The flooding based on polymer gel particles has good prospects for implementation in the Russian oilfields.

Ключевые слова:

полимерное заводнение, полиакриламид, абсорбент, реологические характеристики, ограничение водопритока, выравнивание профиля приемистости.

В связи с постоянным увеличением числа месторождений, выходящих на завершающую стадию эксплуатации, у нефтедобывающих компаний увеличивается запрос на комплексные технологии, способные одновременно обеспечивать увеличение охвата пласта процессом вытеснения и снижение обводненности продукции скважин. При этом традиционной технологией является заводнение вязкими растворами водорастворимого полиакриламида. В статье приведен обзор технологий полимерного заводнения, выявлены преимущества и недостатки данного подхода, в частности, перечислены факторы, способные влиять на реологию полимерного раствора. Несмотря на широкое применение заводнения на основе полиакриламидных гелей, данный метод имеет ряд ограничений, которые в первую очередь связаны с реологическими характеристиками полимера. Неустойчивость реологических характеристик полимера является существенным недостатком, который может стать причиной недостижимости желаемого коэффициента нефтеизвлечения.

Альтернативной для выравнивания профиля приемистости является технология заводнения частицами полимерного геля (ЧПГ) на основе суспензии сшитых частиц полиакриламида. Синтез такого полимера осуществляется заранее и предполагает формирование трехмерной структуры (3D-полимера). С опорой на выполненный литературный обзор приведен сравнительный анализ эффективности технологий на основе водорастворимого полиакриламида и предварительно сшитых полиакриламидных частиц, в результате сделан вывод о преимуществах последних по ряду значимых показателей. Полимеры на основе ЧПГ не меняют реологических характеристик в процессе закачки, при этом частицы могут изменять форму и рваться, обгибая препятствия. Кроме этого, технология позволяет начинать закачку суспензии с меньшим объемом реагента, последовательно подбирая оптимальный granulometricheskii состав в зависимости от конкретных геолого-технологических условий пласта. Заводнение на основе ЧПГ имеет высокие перспективы внедрения на российских нефтяных месторождениях.

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Introduction

Currently, the major part of the remaining oil in place of the Russian oilfields is at the final stages of development. In particular, in the Perm Region, 66.1 and 2.7 % of its oil-production zones are classified (as of the year 2018) as belonging to the third and fourth stages of development. The prevailing of the later stages' producing zones in the oil reserves' structure raises the urge for more effective technologies for improving the oil recovery factor [1, 2]. One of the commonly used and proven methods of improving the oil recovery factor is polymer flooding [3–6]. The use of such water-soluble polymers as polyacrylamides, guar gum and xanthan gum, ethoxylated urethanes, etc. has been well studied and described. However, in the oil industry, the most commonly used polymers are polyacrylamide-based polymer gels.

The polymer flooding technology is used in case of depletion of the oil-producing formation when the interlayer permeability gradient increases. The heterogeneity of the reservoir profile during flooding facilitates the water circulation in the most permeable intervals, while large volumes of oil in place (oil in dead-end areas; oil film covering the rock; oil in capillaries, contained by capillary pressure; remaining oil entrapped by the rock) remain untapped [7-9]. In this case, water can drain through the oil interlayers hardly capturing any oil.

With a viscous polymer solution, more remaining oil in place can be recovered, increasing the oil recovery factor by 5-30% [10, 11]. The classic polymer flooding technology envisages injecting polymer solution amounting to 0.3-0.5 of the reservoir's porous space [10, 12]. This process can take a long time (up to a year or even more), after which the water is injected. In this case, a viscous polymer interlayer between oil and water is built up to slow down and redistribute water flows in the formation, contributing to the larger coverage of the oil-bearing interlayers [13]. The polymer flooding is often recommended for highly viscous oil production [14]. In general, laboratory and oilfield experiments show that polymer flooding may increase oil production by 10-13% [15, 16].

Despite the extensive use of the polyacrylamide gel flooding, this method is subject to certain limitations associated with the rheological characteristics of the polymer. An alternative technology of improving the oil recovery factor and

the injectivity profile alignment is a relatively new method of suspension flooding using preliminary cross-linked polyacrylamide particles. This paper describes the specifics of the traditional polymer flooding highlighting the limitations and risks associated with the technology; it further provides a review of the alternative flooding technology based on preliminary cross-linked polyacrylamide particles; finally, the comparative analysis of the technologies is carried out.

Analysing the efficiency of the standard methodology of the injectivity profile alignment using water-soluble polyacrylamide

The most common technology for the injectivity profile alignment is the polymer flooding involving the injection of polyacrylamide (PAM, Bulk Polyacrylamide Gels). In the remote area of the formation, the polymer gel fills its highly permeable intervals, slowing down and redirecting the water flows into the oil-bearing interlayers with lower permeability. Partially hydrolysed polyacrylamide gel (PHPAM), produced by copolymerization of sodium alkylate and acrylamide, is the most common to use for this purpose.

The extent of PAM hydrolyzation may vary from 25 to 35%. This parameter is responsible for the rheological characteristics of the polymer's water solution. If PAM hydrolyzation rate is below 25 %, the polymer solutions will not feature high viscosity. Excess of the upper hydrolyzation limit causes a material increase of the polymer's sensitivity to the salts contained in the formation fluid. Thus, after the polymer globules are dissolved in the formation water, the hydrolysed residue of the acrylic acid monomer's carboxyl group creates a screen around the globule that prevents the even distribution of the polymer in the fluid. The parameters that influence the ability of PHPAM to change the viscosity of water solutions include the average molecular mass of the polymer chain, PAM concentration and the composition of the formation water, temperature and pressure in the well, as well as the speed and time of the polymer injection [17-20].

The review paper [19] describes the rheological properties of PHPAM solutions. As the shear rate increases, the PHPAM solution behaves as a pseudoplastic fluid with declining viscosity [21]. This phenomenon can be explained by such processes as disentanglement and even distribution of

the polymer globule in the solution. When the critical shear rate is reached, the dilatant properties of the polymer come up, which means that the polymer solution gains viscosity when the shear rate significantly increases. This phenomenon is explained by the formation of additional molecular bonds between the polymer chains due to the evolving polymer packing.

Polyacrylamide solutions feature another rheological peculiarity that is important for polymer flooding. The viscosity of PHPAM solutions increases if a constant shear rate persists for a long time. This peculiarity is known as thixotropy. In total, two types of rheological behaviour of PHPAM are identified: the first type features a slow increase in viscosity to a certain constant value at a low shear rate; in the second case, the polymer abruptly increases viscosity at a high shear rate, and with the lapse of time the viscosity begins to fluctuate materially [20].

When using volumetric PHPAMs, it is important to mind the interaction between the polymer fibres and the formation water salts. Below the critical shear rate, sodium chloride was found to facilitate the reduction of the polymer solution viscosity, while above the critical shear rate, the opposite effect is observed. As the concentration of the monovalent sodium ions in the polymer solution increases, its viscosity goes down, resulting in the rheopectic properties of the polymer solution becoming less pronounced [22-24].

Multivalent cations (Ca^{2+} , Mg^{2+} , etc.) can form both intramolecular and intermolecular complexes with hydrolysed polyacrylamide. The paper [25] presents the researches on the influence of calcium salts concentration on the PHPAM structure. Low concentration of the calcium ions was found to cause intramolecular cross-linking of the polymer in the solution, which results in the polymer flocculation, preventing the solution from getting more viscous. Introduction of additional calcium ions into such a solution creates intermolecular cross-linking of the polymer causing the formation of clusters in the solution. The initially high concentration of the calcium ions facilitates the evolvment of the intermolecular cross-links. In this case, the concentration of calcium ions depends on the extent of the polymer hydrolyzation [25].

The practice of PHPAM cross-linking with Cr^{3+} ions is quite common [26–28]. The paper [28] describes the mechanism of cross-linking

polyacrylamide in the mineralized water with high Cr^{3+} ion content. The high electrolyte content and low polymer and chrome ion concentration were found to cause the domination of the intramolecular cross-linking between the branched parts of the same polymer chain. In case of higher concentration of chrome ions (comparable to the extent of the polymer hydrolyzation), the viscosity of the cross-linked gel is higher as compared to the PHPAM solution with the same concentration.

The literature describes the practice of applying surface-active agents (SAA) as modifiers of the viscous properties of PHPAM. SAAs are capable of interacting with the polymer chains in the solution, thus influencing the rheological properties of the polymer. In particular, evaluation of the influence made by sodium oleate on the PHPAM properties showed that low concentration of SAA causes the PHPAM viscosity to increase [29]. This phenomenon is explained by the links between the molecules of the polymer and SAA. In case of high SAA concentration, the system's viscosity drops abruptly due to the increase of the repulsive force between the micellar aggregates of SAA and the polymer.

Partial hydrophobization of the PHPAM chain has been studied as a tool for stabilizing the rheological properties of the polymer. Thus, the literature provides information about the introduction of additional hydrophobic elements in the polymer backbone [30-32]. When such polymers are dissolved in the water, hydrophobic monomers of the chain are attracted to each other, cross-linking the polymer chains. Such intermolecular interaction increases the viscosity of the solution. If hydrophobic monomers make up a block in the chain, they manifest more solid intermolecular links with similar hydrophobic blocks of another chain. Among the hydrophobic monomers, acrylate and methacrylate derivatives, monomers with alkyl radicals with different chain length, aryl and alkylaryl, fluoroorganic, and zwitterionic monomers can be used. Additionally, the viscosity of hydrophobically modified polyacrylamide gels can be regulated by introducing SAAs, which form intermolecular interactions with hydrophobic sections of the polymer chain [19].

Despite the development of a broad variety of modifications of water-soluble polyacrylamide, it is hard to mitigate the drawbacks of this technology with the acceptable production cost of the technology in mind. Among the main drawbacks of the viscous

polyacrylamide-based polymer solutions, there are: need for specialized expensive equipment for the production of polymer solution on site; difficulty of the polymer viscosity control in the composition; change of viscosity upon injection of the polymer into the well; polymer sensitivity to the metal ions contained in the formation fluids; unpredictability of the rheological characteristics of the polymer in the formation.

Geotechnical conditions of effective flooding using preliminary cross-linked polyacrylamide particles

In the opinion of the authors, a promising alternative to PHPAM is the technology of flooding the oil formations using preliminary cross-linked particles of polyacrylamide, acting like absorbent granules capable of taking up water. Abroad this technology is known as “Preformed Particle Gels”, while the Russian term is translated as “Polymer Gel Particles”. The cross-linked polyacrylamide production consists of the following stages: polymer synthesis (see diagram in the figure), drying, grinding, and division into fractions [33]. The fractional composition of the polymer may vary from micro- to millimetre size. This parameter is selected depending on the reservoir permeability.

The cross-linked polymers are known to absorb water and are therefore classified as superabsorbents. Due to the ability to swell in the water, the polymer particles become soft and elastic but do not dissolve in the water [34]. The absorption capacity of a polymer (the ability to absorb water that is numerically equal to the amount of water in grams 1 g of polymer is capable of absorbing) depends on its composition, temperature of the formation and salts content in the water used to prepare the suspension. The absorption capacity of a polymer depends on the concentration of salt in the water (the higher is the concentration, the lower is the absorption capacity) and the formation temperature (the higher is the temperature, the higher is the absorption capacity). At the same time, polymer gel particles are more stable under the mentioned factors than traditional volumetric gels that significantly lose viscosity as the temperature and water mineralization grow [34]. The main properties of the particle polymer gel are set out in the table.

The technology essentially consists of the on-site production of low-concentrated suspension using polymer gel particles and formation water that is

injected into the injection wells. As the polymer absorbs water and swells, it can flow only through the most permeable interlayers. The polymer plugs that occur in them cause redistribution of fluid flows and involvement of previously idle intervals of the pay zone into the displacement process.

This technology has some advantages compared to the polymer flooding with PAM-based highly viscous solutions. The polymers based on polymer gel particles are resistant to the metal cation contained in the formation water, they do not change their rheological characteristics under high injection speed and do not create the risk of clogging oil-saturated interlayers with polymer clots [34].

Laboratory tests have shown that swollen polymer gel particles may go through the pores and fissures with the flow area of up to 75% of dry particle diameter. For example, if a particle has a diameter of (D_{gel}) of 10 μ m, after swelling it will be able to go through a channel with a diameter (D_{por}) of 7.5 μ m. This can be roughly compared to the permeability of the interlayer of 350 mD.

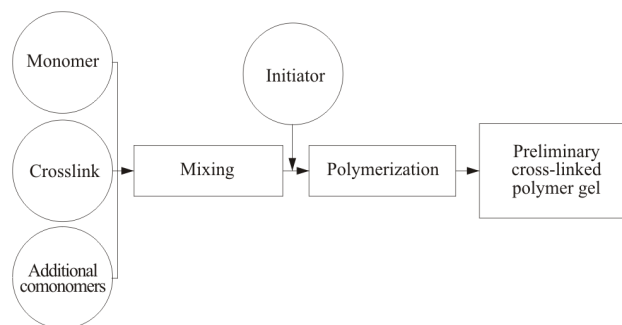


Fig. Cross-linked polyacrylamide particle gel production principle

Principal characteristics of polymer gel particles

Parameter	Characteristic
Size	From 10 μ m to 1 cm
Absorption capacity in the formation water	1-100 g of water per 1 g of polymer
Stability in the salt water	Resistant to any concentrations of salts
Resistance to higher temperatures	1 year at the temperature below 140°C
Durability	Can be regulated by the composition.
Swelling rate	From several minutes to 20 days

The following mechanisms of particle polymer gel impact on the reservoir are possible: deformation of the particle and further movement through the formation; break of the particle and further movement through the formation; jamming in the channels and blocking separate interlayers. Paper [35] describes the fluid flow tests conducted on the terrigenous bulk core. According to the test results, a swollen polymer can penetrate the porous space with a diameter of 18 % of the particle's diameter. A dry polymer particle can go through a pore with the size of 83 % of the particle's diameter [36].

The paper [36] provides the results of the researches aimed at assessing the ability of polymer gel particles to pass through the bulk core porous space. The experiments showed that polymer gel particles can go through the porous space of the core if the ratio of a swollen particle to the channel diameter is less than 15. If the grain size distribution is higher than the reservoir permeability in the bottomhole formation zone, there is a probability that a polymer interlay will appear close to the well edge, considerably hampering the fluid flow.

Production experience of applying the flooding technologies based on preliminary cross-linked polyacrylamide particles

The technology using preliminary cross-linked polyacrylamide particles has been successfully deployed in the leading oil-producing countries (China, the USA, Canada) [35, 37-39]. The global practice polymer gel particles application practice comprised over 4000 well operations as early as in 2013 [35]. In the Russian oilfields, however, it has not been so common so far. Moreover, Russian literature hardly provides any information about the use of polymer gels with limited swelling ratios to modify the well injectivity profile.

The paper [37] has summarized the 20-year world experience of using polymer gel particles technology in various geotechnical conditions of oilfield development: different fluid flow and volumetric properties and reservoir types; temperatures of formations, salt content in the formation water, etc. Thus, as a result of flooding using polymer gel particles for terrigenous reservoirs of the Pushen oilfield (China Sinopec oil company, China), in the conditions of permeability of 121 mD and formation temperature of 107 °C, the water cut of the well production was reduced

from 85 to 70 %. Consequently, daily well production increased from 40 to 60 tons. When the same technology was used at the Xinbei oilfield in China (with the formation temperature of 45 °C and the formation water salt content of 4500 mg/l), the water cut of the well production reduced from 90 to 82 % [37].

Moreover, there are available results of using polymer gel particles technology by PetroChina oil company at terrigenous oil deposits of the Daqing group of oilfields (with the formation temperature of 45°C and the formation water salt content of 4000 mg/l). The polymer gel particles injection into the injection wells caused the reduction of the producing wells' water cut from 95 to 92%, resulting in the daily oil production growth to 5.8 tons. At the same time, for over 2 years additional total growth was observed and totalled 15 000 tons of oil, i.e. additional 113 tons of oil per each ton of used polymer [38].

There is also some experience of using cross-linked polymer-based flooding technologies in the oilfields of Western Siberia, Tatarstan and Kazakhstan. Temposcreen polymer-gel system can be classified as one of them. This reagent is designed to change the well injectivity profile by injecting the water suspension into the formation [40]. When dry, dispersion polyacrylamide gels have the form of powder with the particle size of 0.5-2 mm, increasing to 10 mm in diameter; moreover, the particle size may increase by one thousand times or more. The peculiarity of the technology is the cross-linking of polyacrylamide dry powder with the molar weight of $20 \cdot 10^6$ AMU and degree of hydrolysis of 30% by the ionizing radiation of 10 kGy [41]. The results of using Temposcreen polymer-gel system to align the well injection profile have demonstrated the increase of oil production; the available information indicates 2-8 additional tons of oil depending on the geological structure of the formation and the amount of recoverable oil in place [42]. The "Retin-10" and "Polycar" reagents used by the Russian oil companies are the analogues of Temposcreen [43, 44].

Another known superabsorbent is AK-639 produced by Acrypol LLC, being a part of the visco-elastic and cross-linked polymer systems of the wells injectivity profile alignment. The technology of the well injectivity profile alignment based on this reagent contemplates the injection of 0.5–1.0 % suspension; the polymer swells and

gelates at the temperature of 70 °C when in long contact with water [45].

Conclusion

Summing up, foreign and Russian experience of using the polymer gel particles technology demonstrates its superiority compared to standard water-soluble polyacrylamide-based technologies. The main peculiarities and advantages of particle polymer gel technology can be summarized as follows.

Water-swelling polymer gel particles are significantly more stable to the impact of salts and may change the absorption capacity depending on the mineralization of formation water. Water-soluble polyacrylamide, on the opposite, abruptly loses viscosity or forms clots when interacting with the formation water.

Regular compositions of water-soluble polyacrylamides are unstable at high temperatures. This issue may be resolved by the introduction of special monomers, which causes an increase in the processing cost. The solutions based on polymer gel particles are much more heat-resistant in a broad range of formation temperatures (up to 140°C).

Depending on the polymer molecular structure and geotechnical conditions during the injection process, PAM solutions can change their viscosity to a great extent, often demonstrating pseudoplastic properties. In case of higher salt content in the formation, a polymer can either lose viscosity when interacting with monovalent metals or form clots under high concentration of polyvalent metals. The instability of rheological characteristics of the polymer is a material obstacle on the way to the desired oil recovery factor. The polymer gel particles-based polymers do not change their rheological characteristics during the injection; however, the particles can change form and break while enveloping obstacles.

A well-known advantage of PAM solutions is their ability to easily filter into the formation. When the polymers based on particle polymer gels are used, the fluid flow is usually complicated. For this reason, before using this technology it is necessary to experiment on core models to determine the optimal sizes of their particles. This issue can be dealt, in particular, by using a promising method of X-ray core tomography [46–48].

Additional risks of reducing effects from the PAM technology include difficulties associated with controlling the injection in terms of speed and

volume of the polymer solution and compliance with the solution preparation standard. A suspension based on polymer gel particles is generally stable at high shear rates. However, if the selected particle size distribution is not optimal, in case of polymer gel particles technology it can also cause the build-up of a polymer plug at the reservoir's surface or in the formation pay zone.

The possible options for improving the PAM technology are related to the modification of the polymer structure by introducing additional reagents, which raises the composition cost. In this regard, the technology based on polymer gel particles has a significant advantage as it allows starting the suspension injection with a smaller amount of reagent, successively selecting the optimal particle size distribution depending on the given geotechnical conditions of the formation. In general, the authors conclude that potential polymer solutions based on particle polymer gels need further scientific research and that the technology itself has good prospects of application in the Russian oilfields.

Acknowledgement

The research has been carried out with the support of the Perm Region Administration. Agreement No. S-26/174.7 (MIG-No. 28).

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Please cite this article in English as:

Ketova Yu.A., Bai B., Kazantsev A.L., Galkin S.V. Analysing the efficiency of flooding oil reservoirs with water-soluble polyacrylamide and preliminary cross-linked polyacrylamide particles. *Perm Journal of Petroleum and Mining Engineering*, 2019, vol.19, no.3, pp.251-262. DOI: 10.15593/2224-9923/2019.3.5

Просьба ссылаться на эту статью в русскоязычных источниках следующим образом:

Кетова Ю.А., Бай Б., Казанцев А.Л., Галкин С.В. Анализ эффективности применения заводнения нефтеносных пластов на основе водорастворимого полиакриламида и предварительно сшитых полиакриламидных частиц // Вестник Пермского национального исследовательского политехнического университета. Геология. Нефтегазовое и горное дело. – 2019. – Т.19, №3. – С.251–262. DOI: 10.15593/2224-9923/2019.3.5