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Review

The Comprehensive Overview of Large-Volume Surfactant Slugs Injection for Enhancing Oil Recovery: Status and the Outlook

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Abstract: Despite the development of alternative energy sources, oil and gas still remain the predominant energy sources in most countries in the world. Due to gradual hydrocarbon reserve depletion and the existing downward trend in the production level, there is a need to search for methods and technical approaches to level off the falling rates. Chemically enhanced oil recovery methods (EOR) by surfactant solution injections are one of the possible approaches for addressing this issue in already developed fields. Most often, surfactants are injected together with polymers or alkalis. These technologies are called surfactant–polymer (SP) and alkali–surfactant–polymer (ASP) flooding. Basically, SP and ASP have been distributed in China and Canada. In this article, in addition to these countries, we paid attention to the results of pilot and full-scale tests of SP and ASP in Russia, Hungary, and Oman. This study was a comprehensive overview of laboratory and field tests of surfactant solutions used for oil displacement in SP and ASP technologies. The first part of the article discussed the physical fundamentals of the interaction of oil with surfactants. The second part presented the main chemical reagents used to increase oil recovery. In the third part, we described the main facilities used for the preparation and injection of surfactants. Further, the results of field tests of SP and ASP in the abovementioned countries were considered. In the discussion part, based on the considered results, the main issues and uncertainties were identified, based on which some recommendations were proposed for improving the process of preparation and injection of surfactants to increase oil recovery. In particular, we identified an area of additional laboratory and scientifically practical research. The outcomes of this work will provide a clearer picture of SP and ASP, as well as information about their limitations, current challenges, and potential paths forward for the development of these technologies from an economic and technological point of view.

Keywords: chemical EOR; enhanced oil recovery; surfactant flooding; alkali–surfactant–polymer flooding; surfactant–polymer flooding; chemical reagents; pilot application

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1. Introduction

Nowadays, it is becoming relevant to design and implement methods and technologies aimed at maintaining the level of hydrocarbon production, due to the gradual depletion of oil reserves. There are three main approaches to compensate falling oil production rates. The first one is bringing into development oilfields that have been explored but have not yet been developed. The second approach is the exploration of new hydrocarbon deposits. The last one is to implement methods and technologies for enhancing oil recovery at existing fields [1,2].

The vast majority of hydrocarbon fields being developed are heterogeneous formations with low permeability (<10 mD) or those that contain ultra-highly viscous (>200 mPa·s) oil. From the foregoing and taking into account the global average oil recovery coefficient (currently about 35%) [3], it becomes relevant to design and to improve the technologies aimed at increasing the final oil recovery factor at existing fields [4].

Chemical EOR technologies based on the injection of large-volume chemical slugs into reservoirs have become a promising area for contemporary science. For example, this can include the injection of surfactant solutions such as SP, ASP, and their modifications.

On the grounds that water viscosity increases after polymer addition and the mobility of the aqueous phase is reduced, the mobility ratio between the two phases is leveled off. Therefore, the sweep efficiency is improved, and the displacement efficiency is increased.

The injection of surfactant solutions contributes to a decrease in the interfacial tension (IFT) in the "aqueous solution-oil" (primarily by anionic surfactants) as an alteration in the rock wettability, which intensifies the process of detaching the film oil from the rock granules (primarily by cationic and nonionic surfactants) [5].

Surfactant concentrations in solutions with polymers (SP flooding) achieve a 1–2% reduction. By way of contrast, the addition of alkali (ASP flooding) reduces surfactant concentrations in the range of 0.3–1%. The mechanism for the enhancing the oil recovery of alkalis is the same as using surfactants, so the alkalis enter into a reaction with the acidic components of the oil. Alkali is often added in order to reduce the retention of the main surfactant and improve its emulsification with oil. As a result, organic surfactants (primarily anionic surfactants) are produced, which allow the oil to be washed out of the rock. However, the combination of several modern surfactants at a total concentration below 1% makes it possible to achieve ultra-low interfacial tensions without adding alkali.

Today, there are various modifications of these technologies or their combinations with other EOR methods [6,7]. For example, the mechanisms of the influence on residual oil saturation [8] by foam systems are being investigated. A foam system is a composition that consists of gas (usually CO_2 or N_2) and surfactants, which in this case ensures steady gas movement through the reservoir (as an alternative to SP and ASP) [9]. Another example is nanoparticles, which often are represented by oxides of metals and nonmetals and can be used to influence rock wettability [10,11]. Unfortunately, foam systems or nanoparticles have not been extensively applied due to the insufficient knowledge surrounding them.

In this paper, the basic physical principles, main groups of chemical reagents, and necessary facilities for the preparation and injection of a chemical composition into a reservoir were described based on the literature overview. Moreover, we considered the field application of large-volume surfactant–polymer or alkaline–surfactant–polymer slug injections for these projects, which have been implemented since 2000.

The following issues and ways for scientific development were identified as a result of the literature review:

(1) Today, a pressing problem has become the lack of ready-made solutions for different reservoirs conditions on the international market, including the production of effective anionic surfactants that allow the obtaining of an emulsion of the Winsor III variety.

(2) Additional laboratory studies are necessary for obtaining the dependence between surfactant solutions adsorption and the reservoir properties and thermobaric conditions, namely adsorption reduction studies by special inhibitors or alkalis, and also studies to obtain the dependence between displacement of Winsor III and external conditions such as: temperature, salinity, etc.

(3) The actual problem of the technology has become the development of demulsifiers for the separation of the emulsion that is formed as a result of the reaction of an aqueous surfactant solution and oil. Furthermore, there is a need to develop an approach for its batching in the gathering and processing system.

(4) It is necessary to compare the efficiency of residual oil displacement with surfactants using those aimed at reducing the surface tension at the "rock-oil" boundary and surfactants affecting interfacial tension at the "oil-aqueous solution" boundary.

(5) The comparison of SP and ASP still remains an urgent issue in terms of economic and technological efficiency.

For convenience in navigating the paper, we propose using the following flow chart (Figure 1).

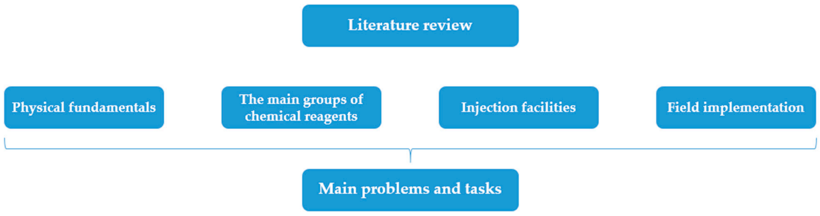

Figure 1. Enlarged research methodology [designed by authors].

2. Physical Fundamentals

Surfactants are used to overcome the immiscibility of water and oil (i.e., surface forces to preventing the production of a single-phase compound of fluids). So, surfactants reduce the interfacial tension between oil and water, thereby stabilizing the compound. In addition, surfactants spontaneously concentrate at the interface or "surface" between immiscible fluids due to their specific chemical structure. A surfactant molecule consists of two main parts. The first one is called lyophilic or hydrophobic, which is attracted to the oil phase. The second part is the lyophobic or hydrophilic part, so it repels oil and attracts the aqueous phase [12].

The schematic definition of wettability in a porous media with an oil/water/rock system illustrating water-wet, oil-wet, and mixed-wet conditions in terms of contact angle and capillary pressure is shown in Figure 2.

The possibility of oil displacement by surfactant solutions is commonly predicted based on their ability to reduce capillary forces, thereby increasing the capillary number. It describes the ratio between viscous and capillary forces [13]:

$$N_C = \frac{v \cdot \mu_w}{\sigma} \tag{1}$$

v—is the flow rate of the displacing liquid, m/s;

μ_w—is the viscosity of the displacing liquid, Pa·s;

σ—is surface tension at the "oil—displacing liquid" boundary, N/m;

The capillary desaturation curve (CDC) is used to determine the effect of the number of capillaries on the residual saturation. The capillary desaturation curve is a graph of the residual saturation of the wetting or non-wetting phase, depending on the number of capillaries.

CDC is an important curve in surfactant formulation because many surfactants can reduce the IFT, which leads to an increase in the number of capillaries and a decrease in residual saturation. Wettability and pore size distribution are two factors that influence the shape of the CDC [14]. As a result, it is assumed that each type of rock in a specified reservoir has distinct CDC slopes and critical points.

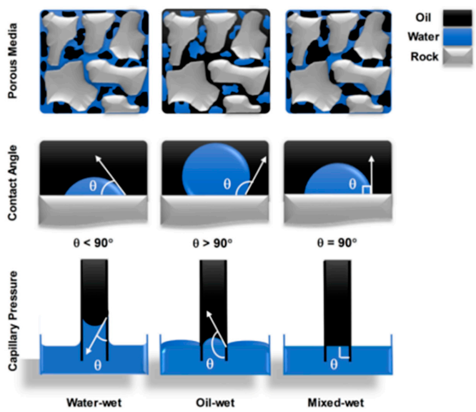

Figure 2. Schematic definition of wettability in porous media with oil/water/rock system illustrating water-wet, oil-wet, and mixed-wet conditions in terms of contact angle and capillary pressure [15].

3. The Main Groups of Chemical Reagents

There are four main groups of surfactants, which are divided by the nature of the hydrophilic group: anionic, cationic, nonionic, and zwitterionic [16]. However, the use of different types of surfactants together in one composition due to the compound effect, in comparison with single surfactant use, is often more effective in reducing interfacial and surface tension. This is because various surfactants allow the solubilization of various components of oil. Thus, the quality features and properties of the outcome composition are altered and expanded by adding different types of surfactants. According to the research [17], this effect is related to the presence of specific interactions between molecules or ions of different types. However, these interactions can either increase or decrease the effect of the compound at the interfaces. As a result, due to the various component compounds of crude oil and applied surfactants, individualized studies are required to clarify the possible effects.

3.1. Anionic Surfactants

These surfactants are often used in chemical flooding projects because of their widespread all over the world, the possibility of altering their properties, and their reasonable cost. This group includes an extremely wide range of molecular structures with different functionalities, but we considered the most commonly used.

3.1.1. Alkyl Aryl Sulfonates

Alkyl aryl sulfonates were first presented to the global market in the 1930s–1940s and they became the main industrial surfactants in 1945. The most significant drawback of these surfactants is their lack of natural biodegradability. Moreover, it has been established that these surfactants are not very stable in conditions of increased water hardness or in the presence of divalent ions (e.g., Mg, Ca). On the other hand, alkyl aryl sulfonates retain their stability in high-temperature formations [16]. Therefore, it is necessary to use alkyl aryl sulfonates in compounds with other surfactants or in compounds with co-solvents to improve their withstanding ability in harsh conditions.

3.1.2. Alkyl Sulfate

The most remarkable examples of representatives of the anionic surfactant family are sodium lauryl sulfate, sodium dodecyl sulfate, and sodium octyl sulfate. In comparison with alkyl aryl sulfonates, alkyl sulfates biodegrade easily. The sulfate group makes the surfactant hydrophilic and water soluble. However, alkyl sulfates become water insoluble in high-temperature conditions because of their temperature sensitivity [18].

3.1.3. Ethoxy Sulfate

Ethoxy sulfates are anionic surfactants with a cloud temperature >100 °C. The presence of sulfonate increases the long-term surfactant stability at a high reservoir temperature in the presence of hardness salts [19].

3.1.4. Alkyl Ethoxy Sulfonates

These anionic surfactants are stable at >80 °C over a broad pH range and a broad water salinity range, even in the presence of divalent ions (Ca, Mg) [20].

3.1.5. Alpha-Olefin Sulfonates

The research results presented in [21] demonstrate that these surfactants perform particularly well in the presence of divalent ions and have a high rate of biodegradation. They are stable over a broad pH range and have a good foaming ability and a good detergency effect in hard water [22–24]. Therefore, these surfactants are widely used as an alternative foaming agent.

3.1.6. Gemini

The paper [25] is one of the most important studies of these surfactants and their properties. In the paper, the effect of Gemini surfactants on interfacial tension was investigated, and their stability under various conditions was also estimated. Thus, these compounds are stable in aqueous solutions even at high temperatures (>85 °C), in high-salinity solutions, and brines without any phase separation or precipitation. In addition, it is possible to achieve an ultra-low IFT, and their adsorption is also low.

Generally, all anionic surfactants are less susceptible to adsorption on negatively charged solid surfaces of a porous media, which characterize sandstone reservoirs [26]. Thus, this makes anionic surfactants attractive enough to mobilize the remaining oil reserves. Hence, anionic surfactants are quite good candidates for increasing displacement efficiency.

To obtain these surfactants, several production processes are used, such as alkylation, alkoxylation, and sulfation [27].

3.2. Cationic Surfactants

We divided these surfactants into two groups: those with carbon–nitrogen bonds and those not containing nitrogen bonds. The first one includes amine salts, among others. There is a reagent produced in Russia called "Katapav" based on them. "Katapav" represents an alkyl dimethyl benzyl ammonium chloride. This reagent is widely available on the market. According to the technical specification, this reagent is incompatible with anionic surfactants. Hence, they are forbidden to be used together. Therefore, it is necessary to investigate the compatibility of the reagent with non-ionic surfactants [28].

Alkyl Benzyl Dimethyl Ammonium Chlorides

These are intended for application as an active base in the production of general-purpose disinfectants, in water-purifying treatments, as functional additives in the production of technical detergents, and also in oil and gas production as biocides of sulfate-reducing bacteria, wetting agents, and corrosion inhibitors.

In addition, there is a Russian corrosion inhibitor "NG-2", which effectively reduces the IFT at the oil–water interface. This substance is composed of oxyethylated amines based on fatty acids C_{10}–C_{17}, containing 6–15 groups (CH_2CN_2O). As an oxyethylated fatty-acid-based amine, the reagent contains primary or secondary fatty acid monoamines [29].

Ethoxylated amines in acidic media are protonated and exhibit anticorrosive, antistatic, bactericidal, and other properties of cationic surfactants. They are characterized by a high resistance to degradation in acidic and alkaline media [30]. The polyester chain of ethoxylated amines gives surfactants resistance to electrolytes and a high water solubility due to the appearance of hydrogen bonds between water and etheric oxygen atoms [31]. However, cationic surfactants are rarely used as reagents to enhance oil recovery [29].

3.3. Nonionic Surfactants

Nonionic surfactants are stable in high-salinity and high-water-hardness conditions. In addition, they are compatible with other types of surfactants.

There are some disadvantages of nonionic surfactants such as their physical state owing to their high viscosity, high cost, and high adsorption levels.

3.3.1. Alkyl Polyglycoside (APG)

APGs are nonionic surfactants and belong to the group of sugar surfactants. Studies on emulsions based on APG were carried out under conditions of high salinity and high temperature in previous research [32]. The chemical structure of glucose leads to a high solubility of APG even under harsh conditions (at a salinity of 180 g/L and at a temperature of 80 °C in the presence of divalent Mg and Ca ions). This behavior of APG leads to low values of interfacial tension at the oil–aqueous solution interface under various conditions [33].

3.3.2. Neodol

Shell researchers [18] have developed the surfactant Neodol for methods of increasing oil recovery. Shell offers more than 30 different nonionic surfactants based on Neodol.

All these surfactants are nonionic and hydrophobic. Shell researchers have recently noted that the addition of other surfactants, such as olefin sulfonate, to the Neodol, forms a mixture that is more effective for influencing residual oil saturation. These surfactants were successfully used up to 57 °C.

3.3.3. Ethoxylate-Monylphenols

In the source [34], the use of surfactants of this type in various conditions was studied. It has been confirmed that this substance retains its stability in carbonate reservoirs at temperatures above 100 °C and at a salinity >200 g/L [19]. Moreover, a mixture of this substance with some cationic surfactants can further increase the stability of the entire formulation [35]. In addition, the researchers suggested that the main mechanism of oil extraction by this surfactant is the change in the wettability of rock.

3.4. Zwitterionic Surfactants

Depending on the media of a solution, this type of surfactant demonstrates anionic or cationic properties (pH or nature of solvent, etc.). Due to the dual nature of surfactants, charged particles can interact with oil and water, and also with some of the minerals of a formation. Sandy and clayish particles have a negative charge, so it could become a problem for cationic surfactant application because of their strong attraction to a negative charge. Therefore, they are not suitable for chemical methods without other surfactant additives; only a minimal amount of surfactant will remain in the liquid phases, which is not enough to effectively reduce the interfacial tension or for residual oil mobilizing [36]. Table 1 shows the main surfactants and their chemical formulas considered in this paper.

Table 1. The types and chemical structures of the surfactants used in EOR methods [18,37].

Surfactant Type	Name	Chemical Structure
Anionic	Alkyl aryl sulfonates Alkyl sulfate	$RO(R_{\varrho}O)_n\ R_{\exists}SO_3–M+$
	Ethoxy sulfonate	$H(OCH_2CH_2)_n–O–SO_2–R$
	Alkyl Ethoxy sulfonates	$ROH–[CH_2–CHO–CH_3]_x–SO-3Na+$
	Alpha-Olefin sulfonates	$R–CH{=}CH–(CH_2)n–SO_3Na$
	Internal olefin sulfonate (IOS)	CH_3-$(CH_2)_6$-CH-$[CH_2]_n$-CH-$(CH_2)_6$-CH_3 + CH_3-$(CH_2)_5$CH=CH-$[CH_2]_m$-CH-$(CH_2)_6$-CH_3
Cationic	Alkyl benzyl dimethyl ammonium chlorides	R-$CH_3CH_3CH_2C_6H_5NCl$
	NG-2 oxyethylated	C_{10}–C_{17}, containing 6–15 groups (CH_2CN_2O)
Nonionic	Alkyl Polyglycoside (APG)	$C_6H_{11}O_5$-O-$(CH_2)_{7\text{-}9}$-CH_3
	Neodol	$RO(CH_2CH_2O)x\ CH_2COO-M+$
	Ethoxylate-monylphenols	$RO–(CH_2CH_2O)_n–H$
	Polyoxyethylene alcohol	$C_nH_{2n+1}(OCH_2CH_2)_m\ OH$
Zwitterionic	Dodecyl betaine	$C_{12}H_{25}N+(CH_3)_2CH_2COO-$
	Lauramidopropyl betaine	$C_{11}H_{25}CONH(CH_2)_{3N}+(CH_3)_2CH_2COO-$

In addition to the surfactants discussed above, there are a huge number of other effective compositions. Specific classes and compositions of surfactants are considered in the next part of the scientific review devoted to the main industrial tests of chemical flooding based on surfactants.

The purpose of the correct selection of surfactant type and composition is to obtain a surfactant–polymer or alkali–surfactant–polymer composition, which will allow the obtaining of the maximum amount of a single-phase (micro) emulsion of oil and water, indicating a low interfacial tension. An alteration in water salinity (a salinity scan) can be used to find the phase behavior corresponding to the best oil displacement. The use of surfactants in water with varying salinity degrees affects the ability of the composition to reduce the interfacial tension [38–40]. In addition, depending on the composition, water salinity, and oil properties, various types of emulsions can be obtained, but it is usually desirable to have a microemulsion (middle phase), also called the Winsor III state (Figure 3).

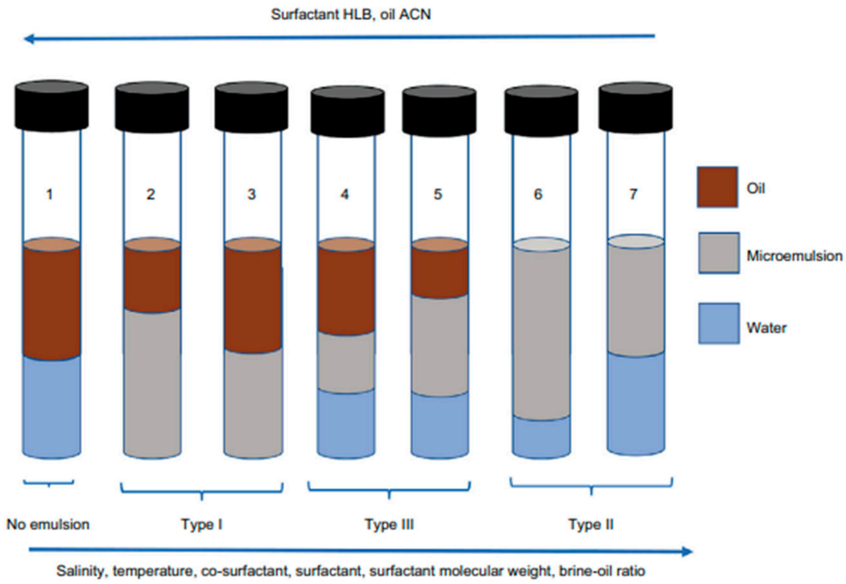

Figure 3. Winsor types [41].

4. Injection Facilities

For polymer flooding implementation, two approaches can be used in relation to the injection facilities, namely using a stationary unit (investment building) and using packaged equipment [36].

Investment building is usually used in cases where polymer flooding is implemented in the entire field or in the vast majority of injection wells. This approach requires significant capital expenses and the approval of necessary permits, but it further reduces the operating costs for maintenance and repair of the injection equipment. In comparison, packaged equipment is used for pilot projects involving from 2 to 10 injection wells or in conditions where the delivery of building materials and construction equipment is limited by external factors. Packaged equipment has some advantages:

- They do not require the redesign of the essential surface infrastructure of a field;
- They can be moved to other fields;
- The do not require additional approval or permits.

Regardless of the approach to equipment designing (stationary or packaged), the flow sheet is common and is shown in Figure 4:

1. Water treatment unit;
2. Polymer solution preparation unit;
3. Polymer storage unit;
4. Nitrogen blanket;
5. Injection unit of (alkali–)surfactant–polymer solution;
6. Surfactant preparation unit;
7. Surfactant storage tank;
8. Alkali storage and batching unit;
9. Water softening and secondary water treatment unit.

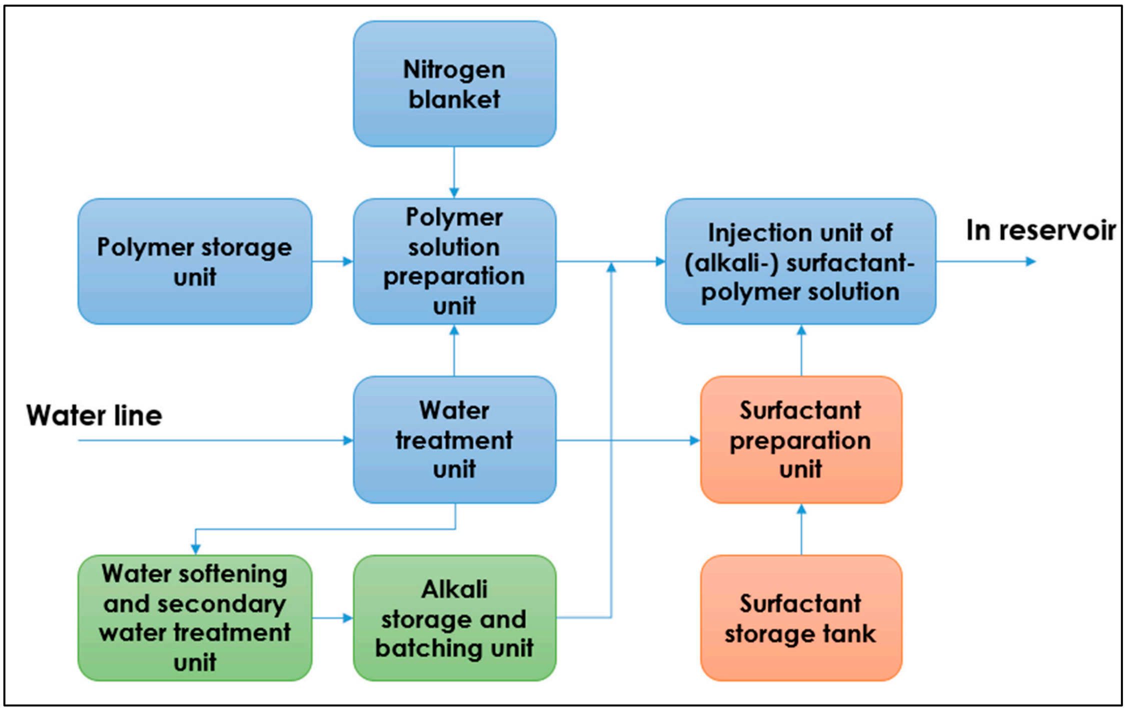

Figure 4. Flow sheet of surfactants injection [designed by authors].

The water treatment unit includes: (1) fine and coarse filters for mechanical impurity removal; (2) an oxygen scavenger; (3) a tank for pure water storage; (4) a rotary vane pump for transferring water to the polymer solution preparation unit; and (5) high-pressure pumps for transferring water to the injection unit.

The polymer solution preparation unit includes: (1) a tank for uploading dry polymer; a unit for the primary and secondary wetting of dry polymer; (3) a maturation tank that contains several chambers with installed mixing machines, which is designed for the homogenization of the polymer mother solution; and (4) a high-pressure pump for supplying the mother solution to the mixing and injection unit.

The injection unit of (alkali–)surfactant–polymer solution includes: (1) flow and pressure regulators; and (2) static mixers for the polymer mother solution and water blending. After the final mixing, the prepared polymer solution is fed into the required number of injection wells.

Throughout the entire technological process, nitrogen is supplied to the buffer tank, to the maturation tank, and to the primary mixing unit from the nitrogen blanket. The nitrogen blanket creates an insulating layer to prevent a contact with oxygen [42].

The surfactant preparation unit includes: (1) a surfactant storage tank; (2) a metering pump for supplying surfactants to a static mixer; and (3) a block for surfactant polymer solution mixing and injecting.

The ambient temperature has a great influence on the shelf life of surfactants. On the grounds that the temperature increases during long-term storage, the hydrolysis process accelerates, so surfactants decompose. In this regard, temperatures between −10 and +20 °C are required for the long-term storage of surfactants. At these temperatures, side effects are also excluded (for example, precipitation, an increase in viscosity, etc.). In the vicinity of the surfactant preparation unit, a surfactant storage tank is located to maintain the continuity of the injection process.

The water softening and secondary water treatment unit includes: (1) a water storage tank; (2) a water softening unit, which may include various units depending on the required degree of water treatment and the contamination level; (3) a prepared water tank; and (4) a pump system for supplying water to the alkali storage and batching unit.

The alkali storage and batching unit includes: (1) an alkali preparation tank; (2) a dispersant; and (3) a metering pump.

5. Outcomes of SP and ASP Flooding Implementation

The first results of laboratory and field tests of ionogenic surfactants used as additives in water flooding were published in the USA in the 1940s–1950s. There were more than 30 surfactant injection (water-soluble and oil-soluble surfactants) pilots on different fields in Russia, but the first one was carried out in 1964 on Arlanskoye field. Aqueous solutions of OP-10 were used.

The oil displacement process by OP-10 aqueous low-concentration solutions is based on a decrease in the surface tension at the oil–water solution boundary from 35–45 to 7–8.5 mN/m and on an alteration in the wetting angle of the quartz plate from 18 to 270. Hence, the wetting tension was reduced by 8–10 times. By the way, BashNIPIneft studies have shown an optimal non-ionic surfactants weight concentration of 0.05–0.1% [43].

5.1. West Salym Field

West Salym field is located in Western Syberia in the Khanty-Mansiysk autonomous district. The field is being developed by Salym Petroleum Development N.V. (SPD). The main specifics of this field are a high reservoir temperature (80 °C) and low oil viscosity (<2 mPa·s).

A maximum oil production of West Salym was observed in 2011, and after that the trend has shown a dramatic fall. Therefore, it was decided to introduce ASP flooding [44]. It should be noticed that the grounds for ASP were prepared in 2008, where some polymers and surfactants were tested under the reservoir conditions of West Salym. The ASP pilot test was launched in 2016 and included four injecting wells, one observation well, and one producing well. The formulation of the chemical reagent was compounded of a blend of two surfactants of the IOS family (0.7 wt.%), 2 wt.% sodium carbonate, 2 wt.% isobutyl alcohol (as a solvent), 0.8 wt.% sodium chloride (as an aqueous solution for salinity altering in the reservoir), and 0.25 wt.% of the polymer Flopaam 3230.

At the beginning of the project an injectivity loss was observed. In this regard, it was decided to stimulate the wells with thermal fracturing [45]. The length of the largest crack did not exceed 12% of the distance to the producing well. Moreover, the pilot results showed a poor separation of the resulting emulsion, which also made the development of an effective emulsion breaker for chemical EOR relevant. Another drawback of the ASP pilot was the strong scale of the downhole equipment due to the chemical in flow to the producing well.

Considering the positive results, an increase in the additional production of about 16% was observed in comparison with the basic production. There was also a decrease in the water cut from 98% to 88%. At the moment, the full-field implementation of ASP is being considered.

As for the various approaches to injecting compositions based on surfactants, everything depends on the goals of the pilot test or large-scale implementation. For example, if the task is to obtain the maximum technological efficiency, then the following sequence of injection of slugs is followed: First of all, a water slug is injected, which contributes to an alteration in the mineralization in the reservoir system, after which the main slug is injected, including surfactants (alkali-surfactants) to mobilize residual oil. Then, a polymer slug is injected, which displaces the mobilized oil. This sequence is illustrated in Figure 5. Of course, the approach to the injection of chemical compositions may differ because it depends on the conditions of the deposits, the stage of development, the well pattern, and other things.

Today, SP projects are being developed in the Bobrikovskian horizon of Romashkino [46] and in the Kharyaga field [47]. There are some requirements for the SP reagent composition in these examples. For example, the SP composition for the Kharyaga reservoir conditions must withstand the high salinity of the reservoir (up to 170 g/L) and the injected water (60–80 g/L), and it also must maintain its properties at a reservoir temperature of 62 °C.

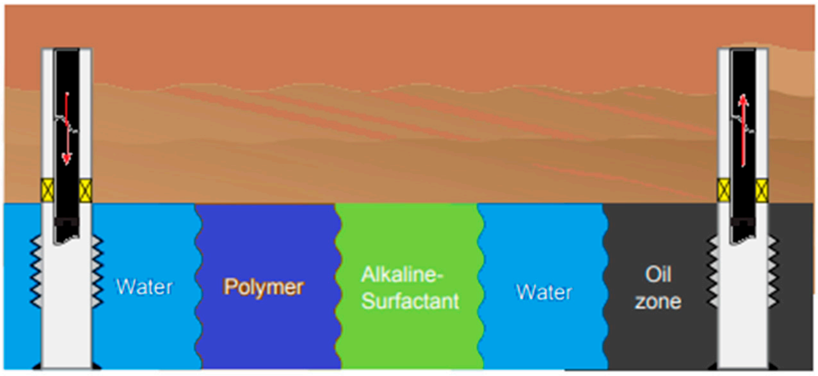

Figure 5. Diagrammatic representation of ASP flooding.

Every field has its own individual features of geological structure, reservoir porosity, and permeability properties. So, the ASP composition for each field is unique. Several papers have been dedicated to the selection process of a chemical composition for SP flooding in carbonate reservoirs in order to enhance oil recovery [48–51]. More than 20 different compositions have been considered, but none were obtained that helped to reduce the interfacial tension to ultra-low values under the conditions of a carbonate reservoir. Thus, the selection of SP chemical formulation for carbonate reservoirs has become a relevant task.

5.2. Algyo Field

It is possible to mention the only SP flooding pilot test conducted in the Algyo field in Hungary [52]. The main distinguishing feature of this field is the high reservoir temperature (98 °C), which was the main problem encountered during the chemical selection. The selection process has been underway since 2001, while the infectivity test was performed in 2013.

The influence of the temperature on the effect of surfactants has been noticed in many scientific papers [53–55]. So, the effect of temperature on surfactants is ambiguous and depends on many factors, for example, the chemical surfactant formulation. Therefore, the temperature effect needs to be studied more fundamentally. The most common temperature effect issue is the loss of surfactant properties (partial or complete precipitation) after passing through the Krafft point (between 30–150 degrees). This parameter is crucial in the surfactant selection process, particularly for high-temperature reservoir flooding. It is also important to notice that this issue requires further study in terms of the formulation selection that allows one to expand the limits of surfactant applications.

The main goals of the pilot were to confirm the efficiency and withstanding ability of the developed surfactant formulation. Thus, a special design of the test formulation was developed, which included the following basic steps. At the beginning, 100 m^3 of water was injected. Then, a surfactant polymer composition was injected. After that, the well was closed for 3 months. In the following step, the pilot well was turned to production to lift the injected surfactant and assess its condition.

Thus, the obtained positive pilot results allowed the study to move to another testing area, which included two injecting and five producing wells. The injection is currently planned to be carried out over 45 months in order to increase the recovery factor. Unfortunately, there is no information about the results of this pilot at the date of this paper's preparation.

5.3. Daqing Field

ASP tests were launched after a successful implementation of polymer flooding in several areas of Daqing. In the paper [56], a significant increase in oil recovery was noticed after ASP flooding.

There is no doubt about the technological efficiency of ASP introduction. However, as a result of ASP flooding introduction, there are some following factors, which are connected with full-field implementation.

Firstly, a major issue is the lack of ability of the industrial output of surfactants near the field. In addition, producing surfactants should be cheap. Secondly, it should be noted that many wells in the study lost their injectivity. This factor needs to be studied. Studies can be provided by a literary review of projects where similar problems have been encountered, as well as by laboratory studies.

In addition to the largest Daqing project, many SP projects have been carried out in other fields in China [57]. The main results are shown in Table 1.

Field tests conducted at more than ten fields in China confirm the technological efficiency, which in some cases has reached an IORF of more than 20% OOIP in a high-permeability reservoir in Liaohe Oilfield. For some projects, there has been a failure to achieve certain targets, namely the injectivity, viscosity, and separation of emulsions. Additionally, a sufficient number of ASP flooding tests were conducted in the fields of China [56]. However, there is still no clear opinion as to which approach is better, i.e., with or without alkali. In one case, the engineers faced problems with an increased adsorption or increased consumption of surfactants. In another case, problems with water preparation or oil demulsification were observed (in the case of using alkali, more resistant emulsions were formed). All these issues require further scientific development. In this article, we did not pay special attention to Chinese projects because there is a lot of information about them, and they deserve a separate publication with an overview of these projects.

5.4. Marmul Field

The following example is the introduction of ASP flooding in the Marmul field, which is situated in the southern part of Oman [58].

Extensive laboratory studies on the chemical formulation selection in this field were run to confirm the ability of the formulation to recover up to 90% of the oil that remained after repeated water flushes, which was approximately equal to 5% of the residual oil saturation.

For the pilot area, an inverted five-spot pattern was selected, which generally included generally seven wells (four producing, one injecting, one observing, and one sampling for continuous development control) with a small distance between them. The selection of this system is usually used in pilot projects, for example, the ASP pilot in the Salym field. The five-spot pattern helps to achieve the main goal of every pilot, which is to obtain the technological effect as soon as possible. When replicating or fully implementing ASP or SP technology, the well-density grid and well pattern have an indirect influence on the overall technological effect. In general, there are no restrictions in relation to the selection of the appropriate well pattern.

The distinguishing feature of this pilot test was the preliminary assessment of residual oil saturation in the investigation area by a single well chemical tracer test (SWCTT). This test is often used in surfactant injection projects where the main goal is to reduce the residual oil saturation. In this regard, the whole process was provided according to the following steps [59]:

(1) Reservoir water injection (to displace mobile oil beyond the intended study radius);
(2) Injecting a tracer and pushing it through a well with water;
(3) Waiting for the reaction for 1–5 days (depending on reservoir conditions). It is necessary to form a secondary tracer through the hydrolysis of the primary one;
(4) Well sampling, where fluid samples are taken from the well at a certain frequency.

As a result of tracer concentration determination in the fluid samples, the dependence between the concentration of tracers and time was obtained. In this case, the oil saturation was a function of the maximum tracer concentration time record [60]. This method allowed a comparison of the residual oil saturation of the reservoir before the injection of chemical reagents and after. As a consequence, the effects of surfactant application was determined. The SWCTT method could become a good tool for the evaluation of the effectiveness of any EOR technology. However, it also had a number of issues that require additional study (i.e., the selection of a tracer for specific field conditions, the interpretation of the obtained data, etc.) [61].

As a result of the preparatory work, a chemical formulation was selected. The composition included 2% sodium carbonate, 0.3% mixture of Enordet surfactants (internal olefin sulfonate) and alcohol sulphate Enordet surfactants, and the polymer Flopaam 3630S.

Thus, the pilot project made it possible to extract an additional >20% in comparison with water flooding. Moreover, according to the data of the surrounding wells, there was a decrease in the water cut of 25–30%. The pilot project was deemed a success, and pre-project work for full-scale implementation is currently underway [58].

5.5. Warner Field

The first full-field implementation of ASP flooding was carried out in the Warner field in Canada, where glauconitic sand has been produced since 2006 [62]. It was an additional project following AP flooding at a nearby field. The characteristics of this deposit are presented in Table 1.

A special composition was developed for this project, which included 0.75% by weight of sodium hydroxide, 0.15% by weight of the anionic surfactant ORS-97HF, and 0.12% by weight of the hydrolysed polyacrylamide SNF Flopaam 3630. Initially, there were 29 producing wells and 11 injecting wells, but later 45 producing and 18 injection wells were added with the help of infill drilling.

After two and a half years of ASP flooding, a gradual transfer from ASP to polymer-only injection was started in October 2008. The first reaction was observed in less than a year (in some wells, after 3 months), but the peak of production rate of 300 m^3/d (~1800 bpd) was achieved in almost 2 years.

Though this application made it possible to extract a significant volume of oil, an increase in the oil volume extracted through ASP flooding remains in question, because a large number of the projects were implemented in the early stages of flooding. The subsequent oil production rate drop is probably explained by a decrease in injectivity, the delayed well response occurring at different times, and well operation problems caused by sedimentation associated with the presence of divalent ions (Ca, Mg) in the water [16]. These substances cause a loss of surfactant properties and surfactant precipitation, which often leads to pore plugging. Anionic systems of surfactants are more sensitive to divalent ions than monovalent ones, especially at low concentrations of surfactants. However, to neutralize these substances in water, several possible solutions have been proposed. Due to economic and technological reasons, it is not always possible to provide a field with the necessary facilities and reagents to neutralize divalent ions. Therefore, the development of a chemical formulation that is capable of working in water effectively in the presence of these substances has become relevant [63,64].

5.6. Mooney Field

The Mooney field is situated in the province of Alberta in Canada [65]. A distinguishing feature of the field is its high oil viscosity, which is not typical for projects where surfactants are used. After the successful introduction of polymer flooding technology for the further study of chemical EOR technologies, the operators decided to apply ASP flooding in another part of the field [66]. The selected chemical formulation consisted of 1.5 wt % Na_2CO_3, 0.15 wt.% surfactant concentration, and 0.22 wt.% of an associative polymer. Water softening, which was provided with the help of a weak acid–cation exchange unit, was required due to the hardness of the reservoir and the injected water.

The composition injection began in September 2011 through 23 injection wells. The main technological data after the injection launch are shown in Figure 6.

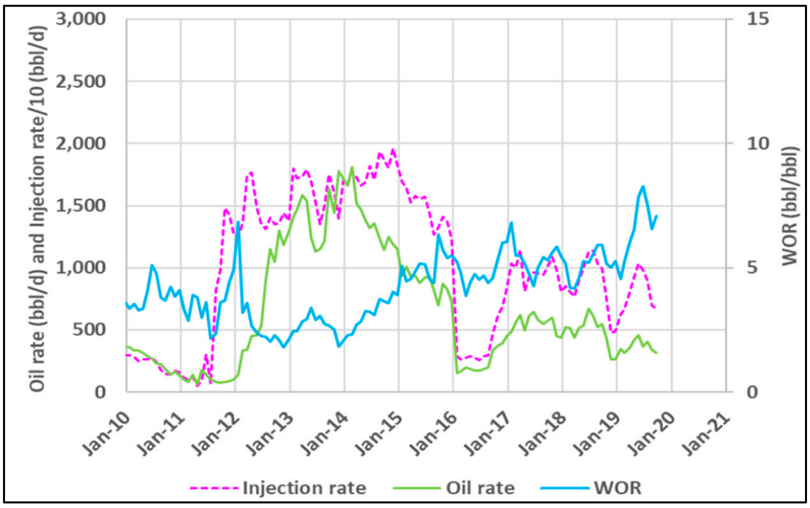

Figure 6. Technological data of ASP flooding implementation in Mooney field [65].

It was difficult to identify the effects of the chemical reagents. There was only a slight decrease in the water cut by the end of 2013. Then, the water cut increased, while the oil production rate began to decline.

The project's halting in 2016 was caused by its high operation costs [66] that were primarily associated with excessive surfactant adsorption. In early 2017, the company decided to resume the project because of the improvement in oil prices.

Table 2 demonstrates the key characteristics of the fields and the brief results of the implementation of SP and ASP flooding in the above-discussed projects. Table 3 includes information on the main chemical reagents used in pilot injections.

Table 2. Key characteristics of fields and brief results of the SP and ASP flooding implementation.

№	Field, Country	Technology	Reservoir Type	Temperature, °C	Oil Viscosity, mPa·s	Salinit, g/L	Permeability, mD	Porosity	Results
1	West Salym Russia [45]	ASP	Sandstone	83	2	15–19	10–100	0.18–0.22	Increase in oil production by 16% in comparison with basic production. Decrease in water cut from 98% to 88%
2	Romashkino Russia [46]	SP	Sandstone	25	30	240	1300	0.23	Pilot is being carried out
3	Kharyaga Russia [47]	SP	Carbonate	62	0.8–1.1	200	300	0.09	Pilot preparation
4	Daqing China [25]	SP	Sandstone	52	12	6	1400	0.26	Increase in oil production from 0.2 mt/y to 4.06 mt/y
5	Zhongyuan China [67]	SP	Not given	80–90	Not given	120	716	Not given	Increase in recovery factor by 13.7%
6	Jilin China [68]	SP	Not given	55	Not given	14	163	Not given	Increase in recovery factor by 14.8%
7	Liaohe China [69]	SP	Not given	55	Not given	3.5	285,9	Not given	Increase in recovery factor by 15.4%
8	Daqing China [25]	ASP	Not given	45	Not given	4.1	500–900	Not given	Increase in recovery factor by 15.0%
9	Changqing China [70]	SP	Not given	51	Not given	12–26	67	Not given	Increase in recovery factor by 15.1%
10	Dagang China [37]	SP	Not given	53	Not given	13.45	675	Not given	Increase in recovery factor by 13.0%
11	Marmul Oman [71]	ASP	Sandstone	46	80	5	1500	Not given	Increase in oil production by 20% in comparison with basic production. Decrease in water cut by 25–30%
12	Algyo Hungary [52]	SP	Sandstone	98	0.64	0.15	70	0.23	Not given

Table 2. *Cont.*

№	Field, Country	Technology	Reservoir Type	Temperature, °C	Oil Viscosity, mPa·s	Salinit, g/L	Permeability, mD	Porosity	Results
13	Warner Canada [62]	ASP	Sandstone	35	58	5.5	2100	0.25	Share of oil in two-phase emulsion has risen from 2–3% to 10–13%. Increase in oil production from 60 m^3/d to 200–300 m^3/d
14	Mooney Canada [65]	ASP	Sandstone	29	300–600	24.5	1500	0.26	Increase in oil production from 100–200 bpd to 2000 bpd. Decrease in water cut from 75% to 55%
15	San Francisco Colombia [72]	ASP	Sandstone	24	10–12	7.6	20	0.17	Increase in oil production by 12–16% in comparison with basic production

Table 3. Chemical formulation of SP and ASP technologies.

№	Field, Country	Technology	Chemical Formulation
1	West Salym Russia [45]	ASP	0.7 wt.% of two surfactants of the IOS family, 2 wt.% sodium carbonate, 2 wt.% isobutyl alcohol, 0.8 wt.% sodium chloride and 0.25 wt.% of the polymer Flopaam 3230.
2	Romashkino Russia [46]	SP	Surfactant by MOL + Flopaam 5115 VHM
4	Daqing China [25]	SP	0.2 wt.% amphoteric (HLW) surfactant + 0.25 wt.% HPAM
5	Zhongyuan China [67]	SP	Not given
6	Jilin China [68]	SP	0.2wt.% PS surfactant + 0.2wt.% HPAM
7	Liaohe China [69]	SP	0.25 wt.% amphoteric surfactant + 0.16 wt.% HPAM
8	Daqing China [25]	ASP	Not given
9	Changqing China [70]	SP	0.12 wt.% amphoteric + anionic + non-ionic + 0.15wt.% HPAM
10	Dagang China [37]	SP	0.25wt.% PS + co-surfactant + 0.15wt.% HPAM
11	Marmul Oman [71]	ASP	2 wt.% sodium carbonate, 0.3 wt.% mixture of Enordet surfactants and alcohol sulphate Enordet surfactant, and also the polymer Flopaam 3630S
12	Algyo Hungary [52]	SP	Surfactant by MOL
13	Warner Canada [62]	ASP	0.75 wt.% sodium hydroxide, 0.15 wt.% ORS-97HF, 0.12 wt.% of the polymer Flopaam 3630 by SNF
14	Mooney Canada [65]	ASP	1.5 wt.% sodium carbonate, 0.15 wt.% surfactant, 0.22 wt.% of an associative polymer
15	San Francisco Colombia [72]	ASP	Not given

6. Results and Discussion

As a result of the review, the following requirements for surfactants were put forward for achieving the maximum effect.

The chemical (resistance to hydrolysis and precipitation) and thermal stability of a surfactant must be maintained during composition preparation, during injection through the well, and during placement in the reservoir.

The solubility of a surfactant is considered satisfactory if transparent or translucent solutions are formed from injected or in-reservoir water at the reservoir conditions.

The stability of a surfactant is considered satisfactory in the presence of hardness salts provided that precipitation or separation into phases is not observed.

The presence of surfactants should reduce the IFT at the "aqueous surfactant-oil" boundary to a value of at least 0.01 mN/m at concentrations not exceeding 1%. A lot of researchers have noted the need to achieve this indicator [46,73,74], as it reflects the possibility of obtaining a medium-phase microemulsion of Winsor III.

Surfactants must be compatible with rock, and undesirable precipitation must not be observed.

Surfactants must be compatible with polymers and (or) alkali compounds, providing that the composition design requires their presence.

Dynamic adsorption needs to be less than 0.5–1 mg/g of rock in reservoir conditions. The specified range is primarily determined by the economic components of the project [75]. At higher values, the costs of injecting the sacrificial agent cannot be offset by additional oil production, as shown in the following works [76,77]. Therefore, studies related to the mechanisms and methods of reducing the adsorption of surfactants on the rock are of particular relevance. In this regard, further research may be devoted to the study of adsorption inhibitors or injection approaches (for example, pre-injection of a less expensive surfactant to saturate adsorption centers).

Based on the modern ideas about reservoir processes during the injection of surfactant solutions, a key issue was noticed, which consisted of uncontrolled losses of active reagents, primarily associated with the following processes:

(1) Adsorption on the rock;
(2) Chemical, thermal, biological, and mechanical destruction;
(3) The redistribution of the surfactants to the oil;
(4) Precipitation as a result of interaction with polyvalent ions (Ca, Mg) in the reservoir water.

The influence of the last two processes can be eliminated by an ordinary selection of the reagents. However, the management of the adsorption processes has become a relevant issue.

Specific approaches are required to reduce the adsorption:

(1) The correct selection of the average molecular weight of surfactants;
(2) The alteration of the composition pH;
(3) The preliminary suppression of adsorption centres on the rock due to the injection of "sacrificial" reagents.

7. Conclusions

(1) Technologies surrounding large-volume injections based on surfactants have a high technological potential.

(2) Today, a pressing problem has become the lack of ready-made solutions for different reservoir conditions in the international market, including the production of effective anionic surfactants that allow the obtaining of an emulsion of the Winsor III variety.

(3) Additional laboratory studies are necessary to obtain the dependence between surfactant solution adsorption and a reservoir's properties and thermobaric conditions. Adsorption reduction studies using special inhibitors or alkalis and studies to obtain the dependence between the displacement of Winsor III and external conditions such as: temperature, salinity, etc., are also required.

(4) The actual problem of the technology has become the development of demulsifiers for emulsion separation that is formed as a result of the reaction of an aqueous surfactant solution and oil. Furthermore, there is a need to develop an approach to its batching in the gathering and processing system.

(5) It is necessary to compare the efficiency of residual oil displacement observed when using surfactants that are aimed at reducing the surface tension at the "rock-oil" boundary and surfactants affecting the interfacial tension at the "oil-aqueous solution" boundary.

(6) The comparison of SP and ASP still remains an urgent issue in terms of economic and technological efficiency.

(7) Our future studies will be focused on injection approaches. We are going to study the pre-injection mechanisms of a less expensive surfactant to saturate adsorption centers.

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