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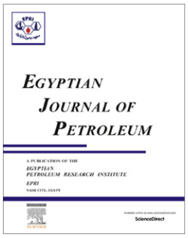

REVIEW

Hydrophobically associated polymers for wettability alteration and enhanced oil recovery – Article review

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Abstract Crude oil and other petroleum products are crucial to the global economy today due to increasing energy demand approximately (∼1.5%) per year and significant oil remaining after primary and secondary oil recovery (∼45–55% of original oil in place, OOIP), which accelerates the development of enhanced oil recovery (EOR) technologies to maximize the recovered oil amount by non-conventional methods as polymer flooding. This review discusses enhanced oil recovery methods specially polymer flooding techniques and their effects on rock wettability alteration.

Contents

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1. Introduction

1.1. Background

Crude oil is the most critical energy source in the world, especially for transportation, provision of heat and light as there has not been a sufficient energy source to replace crude oil has broadly integrated (i.e. Today's energy needs are met in large part by crude oil). Petroleum products are crucial to the global economy today due to increasing energy demand approximately 1.5% per year [1] associated with population growth and improving life styles, limited proven oil reserves (i.e. shortage of current oil resources), declining oil production since 1995, difficulties in finding a new oil fields, non-productive primary and secondary recovery, significant oil remaining after secondary recovery (~45–55% of original oil in place, OOIP), and forecasts for tightening oil supply which drive the need to maximize the extraction of the original oil in-place for every reservoir, and accelerating the development of enhanced oil recovery (EOR) technologies in the high-temperature high-pressure (HTHP) offshore reservoirs. Although the EOR methods have been intensively investigated and implemented worldwide for several decades but until now, after all best efforts, only about 45% of the original oil in place can be recovered by the primary and secondary recovery processes [2], while the rest of oil is still trapped in reservoir pores. The U.S Department of Energy estimates that nearly 377 billion barrels of discovered oil are left behind after conventional primary and secondary production techniques have been employed [3]. By estimation of nanotechnologists and petroleum experts, using nanomaterials in the oil and gas exploration and production, a market of multibillion dollar totally can be created [4].

1.2. Objectives

Crude oil production from sandstone and carbonate rock formations occurs in three distinct phases [5]. Primary oil recovery combines the native energy of the reservoir, which typically recovers about ~15% OOIP [6]. In secondary oil recovery, as the reservoir loses its energy, an external fluid such as water or gas, is injected into the reservoir to maintain reservoir pressure and extend its lifetime recovering an additional ~30% OOIP. After water flooding, there is a significant oil amount ~55% OOIP still trapped in the reservoir rock pores due to both microscopic and macroscopic factors and it cannot be further removed without the use of chemical, thermal or gas injection processes [1].

To increase the oil recovery efficiency in oil-wet reservoirs (unswept regions), different techniques have to be pursued [7];

(A). Improving volumetric sweeping efficiency by adjusting the oil/water mobility ratio through polymers flooding agents which increase displacing fluid viscosity [4], thus increasing produced crude oil amount.
(B). Altering the wettability of porous reservoir rock surfaces to more water wet (i.e. by letting the value of contact angle $\theta \leqslant 90$) [8].
(C). Increase the oil displacement effectiveness by overcoming the capillary barrier through viscous and gravitational forces. Reduction of capillary pressure forces can be achieved by surfactants flooding to lower the oil–water interfacial tension (IFT), to ultra low values $\sim 10^{-3}$ dyne/cm [9].

1.3. Polymeric Surfactants (Surfmers)

Polymeric surfactants are one kind of functional surfactants, which not only have amphiphilic structure composed of hydrophobic tail and hydrophilic head group [10], but also contain polymerizable vinyl double bonds [11] in their molecular architecture resulting in novel physicochemical properties distinct from conventional surfactants [12] such as;

(A). Analogous to common surfactants, they have surface activity; similar to general vinyl monomers, so they can be initiated and polymerized.
(B). Due to amphipathic property and polymerizability of surfmers they can be used to synthesize inorganic/organic nanocomposite, can be applied to emulsion polymerization as polymerizable emulsifiers, to surface modification of solid substances, to synthesis of novel water-soluble hydrophobically associating polymers with strong thickening properties [13] so, they have great significance in enhanced oil recovery [14].
(C). Offer potential for developing hybrid nanosized reaction and templating media with constrained geometries. Moreover surfmer can be directly used as hydrophobic monomer to copolymerize with acrylamide (AM) forming hydrophobically associative polyacrylamide (HAPAM), which has been widely used in enhanced oil recovery, drilling fluids, coats and paintings [15].

Typical polymerizable groups which have been exploited are vinyl, allyl, acrylate, methacrylate, styryl and acrylamide [16]. Position of the polymerizable group either "H-type" where, polymerizable group located in the hydrophilic head group, or "T-type" where, polymerizable group located in the hydrophobic tail have a profound effect on surfactant self-assembly and properties [17,18].

2. Polymer flooding survey

Chemical flooding of oil reservoirs is one of the most successful methods to enhance oil recovery from depleted reservoirs at low pressure after secondary recovery (water flooding). A lot of papers and reviews, both laboratory work and field tests have been published on this subject since the first work by Marathon oil company in the early 1960s [19]. Even though enormous effort by oil companies, university, and government researchers during the 1970s and 1980s increased our knowledge about the chemical flooding process, it was more or less accepted or concluded by the oil companies at the end of the 1980s that the method was not economical, or the economical and technical risk was too high with the present oil prices. Surfactants and polymers are the principal components used in chemical flooding, so chemical flooding is also denoted as micellar/polymer flooding. In this survey, up-to-date literatures on various aspects considered in chemical enhanced oil recovery are reviewed in-depth to illustrate their advantages and drawbacks with emphasis on wettability alteration.

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In polymer flooding processes the concentration of polymer ranges from 500 to 2000 mgL^{-1}. The volume of the polymer solution injected may be 50% PV, depending on the process design [20]. Designing and developing a series of efficient polymer structure is significant for oil and gas exploitation. Therefore, a remarkable number of hydrophobically associated water soluble polymers, including N-(4-ethyl) phenyl acrylamide and acrylamide (AM) copolymer [21] methyl acrylic acid-2-dimethylamino ethyl acrylate and methyltert-butyl ester copolymer [22] methyl acrylic acid, ethyl acrylate and poly (ethylene oxide) (PEO) copolymer; Hydrophobically modified alkali-soluble emulsion polymers[23] and PEO99–poly(propylene oxide)67-PEO99 (F127) multiblock copolymers [24] have been developed since the 1980s [25]. Polymer flooding has been the most used EOR chemical method in both sandstone and carbonate reservoirs. To date, more than 290 polymer field projects have been referenced or reported in the literature. Studies of more than 200 polymer floods reported average polymer injection of 19 to 150 lb/acre-ft and concentrations ranging from and 50 to 3700 ppm, respectively [26]. While related additional oil recoveries vary from 0 up to 18% of OOIP [26,27], reported some advantages in using anionic polyacrylamide/acrylic acid (PAM/AA) made by copolymerization in tests carried out on sand pack and native cores from the Richfield East Dome Unit (REDU) in California. Lower retention and slightly improved oil recoveries were cited.

Flooding tests were conducted in heterogeneous porous media showed that pre injection of polymer could result in better flooding efficiency [28]. Furthermore, polymer pre injection had no effects on oil displacement characteristics of the micellar fluid and appeared to only reduce the surfactant adsorption on the rock for the polymer micelle system studied.

Poly (alpha-alkoxy) acrylamides claimed improved stability in brine solutions [29]. Similar claims prepared N-substituted PAM/AA via ethoxylation [30,31].

Also, terpolymers of acrylamide, acrylonitrile and acrylic acid were prepared [32], the latter optionally alkoxylated with ethylene oxide [33].

Copolymerization of sulfonated monomers such as 2-acrylamido-2-methylpropane sulfonic acid (AMPS) with acrylamide monomers were prepared [34,35]. Molecular weights obtained for such copolymers were not as high as the acrylic acid counter parts. The AMPS copolymers did provide a somewhat improved calcium ion tolerance.

Some studies reported about the synthesis of polyacrylamide containing imide rings and concluded that they are less susceptible to alkaline hydrolysis than PAM [36]. Moreover, viscosity retention in brine for hydrolyzed PAM/AA versus copolymerized PAM/AA were evaluated and found little difference in the two, claiming that shear resistance of the hydrolyzed PAM/AA was superior to the copolymers [37].

Some studies reported that surfactant adsorption is minimized in the presence of polymer thus the use of a surfactant-polymer flood could be highly favorable and justified [38].

Other efforts discuss the limitations of the use of polymer floods and mention that being introduced earlier in the life of a water flood is a better option [39]. They also report incremental oil recoveries in the order of 5% on average. They note also that very few field applications have used biopolymers like Xanthan gum in their floods. From these previous studies, an important consideration therefore in the use of polymers is the point in the life of a water flood at which polymer injection is initiated. Also, worthy of note is the fact that biopolymers like Xanthan have not been widely used. Moreover when they are used in powdered form, gel formation could hamper their performance.

Some trials discuss the concept of “Low Tension Polymer Flood” (LTPF), and conclude that; the flood in the first instance was conducted by co injection of the surfactant and polymer and, due to chromatographic effects, the polymer moved ahead of the surfactant. In such application, mobility of water at the front will be reduced and the activity of the surfactant will be enhanced [40].

Other studies discuss the field application of polymer floods and conclude that when applied after the reservoir has been extensively flooded by other means, polymer floods have been unsuccessful [41]. Another observation they make is that in reservoirs of low average permeability, injectivity of the floods greatly reduced with the addition of a polymer and resulted in poor performance.

recently some authors reported about the synthesis of a novel surfmers (H-type) by the reaction of a 1- vinyl imidazole as a polymeric moiety containing double bond and 4- Dodecyl benzene sulfonic acid surfactant, then hydrophobically associating polyacrylamide (HAPAM) prepared by free radical emulsion polymerization of acrylamide (AM) monomer, divinyl sulfone as hydrophobic crosslinked moiety and surfmers, to chemically anchor a surfmer and hydrophobic crosslinker moiety onto the hydrophilic back bone of acrylamide chain [42,43]. After that, the modified nanocomposite (HAPAM-SiO_2) was synthesized by reaction of HAPAM copolymer with 3-amino propyl) triethoxysilane. The structure of the synthesized copolymer and nanocomposite was analyzed by means of FTIR, ^{1}H-NMR, ^{13}C-NMR, TEM, SEM, XRD and DSC. Flooding experiments carried out on one dimensional sandstone model where, recovery factor reach to nearly 48% and 60% of residual oil saturation (%S_{Or}) in case of HAPAM and HAPAM-SiO_2 respectively at a concentration of 2000 mgL^{-1}.

3. Principle and mechanism of polymer flooding for enhanced oil recovery (EOR)

Polymer flooding can increase recovery up to 5–30% OOIP [44]. Polymer flooding process involves injection of polymer “slug” followed by continued long-term water flooding to drive the polymer slug and the oil bank in front of it toward the production wells as shown in Fig. 1. Based on the principle of mobility ratio, water-soluble polymer reduces water mobility by two mechanisms: (1) increasing the viscosity of the water phase (2) reducing the relative permeability of water to the porous rock by adsorption/retention of the polymer in the rock pore throats [45] and thereby creating a more efficient and uniform front to displace unswept oil from the reservoir (i.e. the mobility ratio (M) is inversely proportional to the water viscosity). With a reduced mobility ratio, the sweep efficiency is increased and, as a consequence, oil recovery is enhanced [46].

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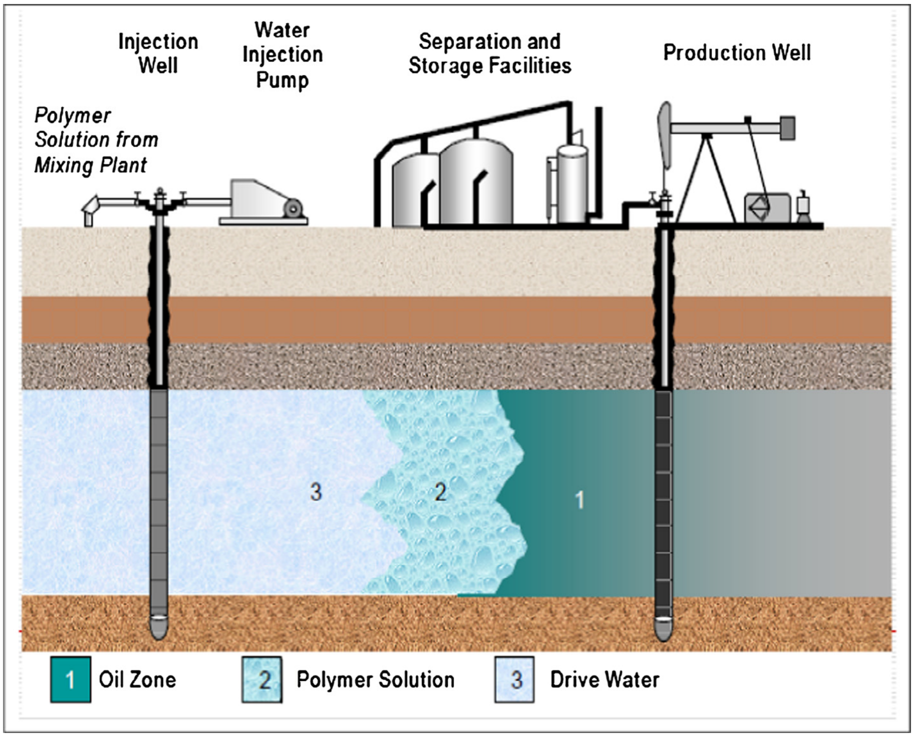

Figure 1 Polymer flooding mechanism (International Energy Outlook, September, 2008).

4. Hydrophobically associated polyacrylamide polymers (HAPAM)

These polymer classes have attracted much attention on both academic and industrial laboratories for polymer flooding in enhanced oil recovery [47,48] because of their unique structures and properties, including their thickening properties, shear thinning, and anti polyelectrolyte behavior which has been widely investigated in oil chemistry additives such as mobility control agents and rheology modifiers [49]. In addition to enhanced thermal stability, relative permeability modifiers, sweeping efficiency, salt-tolerance behavior and high viscosity properties for IOR or EOR applications [50]. These polymers were synthesized by modification of partially hydrolyzed polyacrylamide (HPAM) through grafting or incorporating hydrophobic chain cross-linking segments onto their hydrophilic main chain [51,52] or by copolymerization of hydrophilic and hydrophobic monomers [53]. They are considered as promised EOR candidates for polymer flooding in high salinity reservoirs, owing to their unique characteristics [54] which can be summarized as follow;

A. In aqueous solutions, above a critical association concentration (C^*), their hydrophobic groups develop intermolecular hydrophobic associations in nanodomains, leading to building up of a 3D-transient network structure [55] in high ionic strength medium [56] so, providing excellent viscosity building capacity [48,52,57], remarkable rheological properties and better stability with respect to salts than the unmodified HPAM precursors [58].
B. Reduce interfacial tension at solid/liquid interface, since hydrophobic moieties associate forming aggregates or micelles.
C. Shows an unusual adsorption isotherm [59] so, can be considered as a wettability modifier.
D. Does not undergo mechanical degradation under high shear stress such as those encountered in pumps and near the well bore area, since the physical links between chains are disrupted before any irreversible degradation occurs, also they reform and retain their viscosity upon shear decreasing[60].
E. High resistance to physicochemical conditions (temperature, pH, and ion content) prevailing around the wells, so considered a prospective EOR candidate as thickeners or rheology modifiers in high temperature, high pressure reservoirs [61–63], reservoir stimulation [64] and tertiary oil recovery [65].

5. Reservoir wettability

Wettability defined as ‘‘the tendency of one fluid (wetting fluid) to spread on or adhere to a solid surface in the presence of another immiscible fluid (non-wetting fluid)” [66]. Reservoir wettability is an important and elusive petrophysical parameter in all types of core analysis, which affects saturation and enhanced oil recovery processes [67]. There is a Conesus in petroleum engineering that preferentially water-wet cores flood more efficiently than oil-wet cores; since, more oil is recovered from water-wet cores in the early flooding stages than from oil-wet cores (Jiang et al. [24]). Enhanced oil recovery by wettability alteration of the reservoir rock is the main subject in this work. From 1990 until today the concept of wettability, its impact on oil recovery, and the understanding of alteration mechanisms, had significant attention in the oil industry. Changes in wettability induce changes in capillary pressure with respect to rate and recovery. This has been reported in numerous publications [68–73]. A key parameter, also strongly dependent on wettability, is the residual oil saturation after water flooding. Literature generally reports that the residual oil saturation is a function of wettability, oil and water viscosity, formation water properties (salinity), pore network, and permeability. Some literature reviews favored surfactants, while others found that thermal method will work better in altering wettability and that it is less expensive compared to the chemical methods. Many experimental investigations on the impact of wettability have been conducted and several excellent review literatures are available [74–78].

Others [79] investigated the relationship between wettability and waterflood oil recovery in a series of Berea cores. Their study showed that oil recovery by water flooding initially increased and then decreased as the wettability changed from strongly water-wet to oil-wet [80].

The effect of wettability on oil recovery, using chemical additives that change the surface properties of natural samples while keeping interfacial tension and viscosity constant were studied [81]. The results obtained from their experiments using sandstone samples treated with water soluble potassium methyl siliconate, showed that residual oil saturation (S_{or}) was reduced from 0.4 to 0.3 and 0.1 [82]. Wettability alteration of Berea sandstone using Prudhoe Bay crude oil and synthetic formation brine was also reported [74].

Impact of wettability alteration on two-phase flow characteristics with a network extracted from a sample of Bentheimer sandstone was studied [83]. The results showed that as the system became less water-wet, the residual oil saturation initially decreased but increased dramatically at the transition from water to oil-wet conditions and then decreased to a minimum in oil-wet systems.

Surfactant-induced wettability alteration process appears beneficial for field implementation in oil-wet reservoirs [84]. In these reservoirs, the surfactants can induce wettability alterations to either less oil-wet or less water-wet states, thus improving oil recovery. In initially water-wet reservoirs, the surfactant-induced wettability alteration process is beneficial only if the surfactant induces either mixed wettability or intermediate wettability.

A systematic approach to investigate the oil recovery in chalk as a function of wettability was also presented [85].

Recently a published study [42,43] reported about a novel copolymer and its modified nanocomposite which can alter the wettability of sandstone rock from oil-wet to water-wet, and consequently enhance oil recovery factor.

6. Conclusion

Hydrophobically associated polymers and their modified nanocomposites considered as one of the most modern water flooding techniques through different EOR technologies. The recovery factor is strongly related to petrophysical and geological properties of reservoir in addition to solution and rheological properties of applied polymer. Most of published literature reported about increasing recovery factor by surfactant flooding, however nowadays the recent articles reported about wettability alteration through hydrophobically associated polymers grafted by surfmers, where the results were promised.

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