Recent advancement of hybrid materials used in chemical enhanced oil recovery (CEOR): A review

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Abstract. Depletion of natural oil reserves has forced oil industries to focus on tertiary recovery methods to extract residual oil after exhausting the primary and secondary methods. Among the Enhance Oil Recovery (EOR) technologies, Chemical EOR (CEOR) is gaining popularity. Despite research efforts to increase the recovery using CEOR, increasing complexity in extraction methods are encountered. With changes in reservoir conditions (high temperature, pressure and salinity) and crude oil properties, existing chemicals used in CEOR, such as alkali, polymers and surfactants do not function desirably. These conditions have detrimental effects on the performance of EOR chemicals, like precipitation, degradation, etc. Development and utilization of effective EOR hybrids such as surfactant-polymer, polymer-nanomaterial, surfactant-nanomaterial and polymer-surfactant-nanomaterial had prevailed the effects of harsh reservoir conditions, and their applications in oil fields in recent years have increased the success of EOR. The synergistic effects between the hybrid components play major roles in improving the properties that could withstand the effect of extreme reservoir conditions and changes in crude oil properties. Therefore, this paper is aimed at reviewing recent advances in CEOR hybrid technologies, and discusses the basic concept, applications, advancement and limitations of different hybrid materials used in CEOR processes.

Keywords Reservoir conditions, hybrid, chemical enhanced oil recovery.

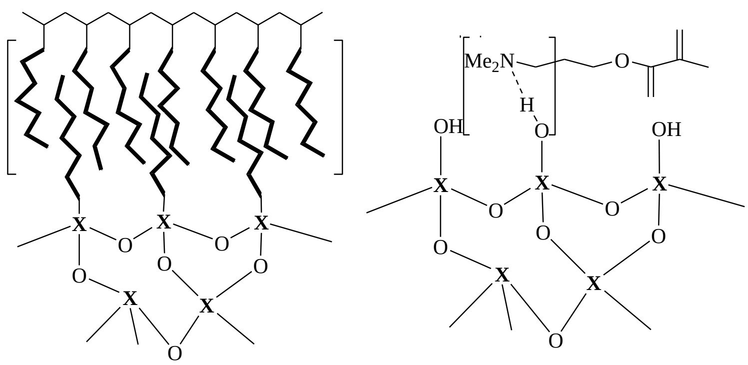
1. Introduction

It has been estimated that two-third of the original oil in place (OOIP) is left in the reservoir after exhausting the primary and secondary methods. Among the factors responsible for oil remaining in the rock pores are the effects of wettability, negative capillary force, and interfacial/surface tension between different phases [1-3]. Despite the discovery of new resources, recently, the United States Department of Energy reported that the world's consumption of crude oil has reached approximately 95 million oil barrels per day, and the domestic crude oil production is not sufficient enough to meet the requirement for energy [4-5]. This indicates over dependency on oil for daily activities. Thus, it has become necessary to initiate and develop other production strategies to recover the last drop of OOIP in the natural reserves. Enhanced oil recovery (EOR) technologies have been primarily designed to increase the oil production, and to bring back the lives of depleted oil fields in order to meet the world's energy demand. Among the EOR technologies, CEOR is the most effective and has the potential to target the residual oil compared to non-chemical methods like the *in-situ* and microbial flooding. The ability of this method to minimize or overcome the wetting and interfacial/surface forces, has led to it receiving attention for applications in oil fields [6-10]. CEOR requires the injection of specific chemical fluids (usually in solutions) such as alkali, surfactants and polymers [8-9]. Each of these chemicals has a specific function when used as a flooding agent, for instance, surfactants usually promote the recovery of trapped oil in the reservoir by reducing oil/water interfacial tension [11]. The polymer is added to increase the viscosity of water, thereby reducing oil/water mobility ratio, which results in improved sweep efficiency [12]. The alkali reacts with the acid present in the oil and reduces the adsorption rate of surfactants on the rock surface [10]. With the changes in reservoir conditions like high temperature, pressure, salinity, and crude properties, flooding with either surfactant, alkali or polymer alone does not give the optimum recovery, because these conditions have detrimental effects on their individual performance (like precipitation, degradation etc.) [13-15]. However, this problem could be prevailed if hybrid materials comprising of surfactant-polymer, polymer-nanomaterial, surfactant-nanomaterial or polymer-surfactant-nanomaterial are used as CEOR agents. The synergistic effects between the hybrid components have increased the success of EOR operations by improving the rheological properties, as well as increasing the thermal and salinity resistance of CEOR hybrids [16]. The ability of CEOR hybrids to withstand the effects of changing crude properties is also associated with the combining effects of the hybrid components [16]. Therefore, in this paper, we discuss the applications, recent advancements, and limitations of different hybrid materials used in CEOR processes.

2. Basic concept

2.1 Hybrid materials

Hybrid materials contain two or more composite materials (mostly organic and inorganic) in a definite molecular ratio. The properties of hybrid materials differ from the individual properties of its corresponding components. In many cases, improved properties are usually due to the interaction of the hybrid materials, because one component provides bonding while the other imparts mechanical strength [17-18]. Depending upon the nature of hybrid components involved, hybrids can exist as liquid or solid phases and are categorized as *class I* (figure 1a and b) with weak chemical attractions (hydrogen bonding, Van der Waal force, etc.) and *class II* (figure 1c and d), which have strong interactions [18].



a. Vander waals

b. Hydrogen bond

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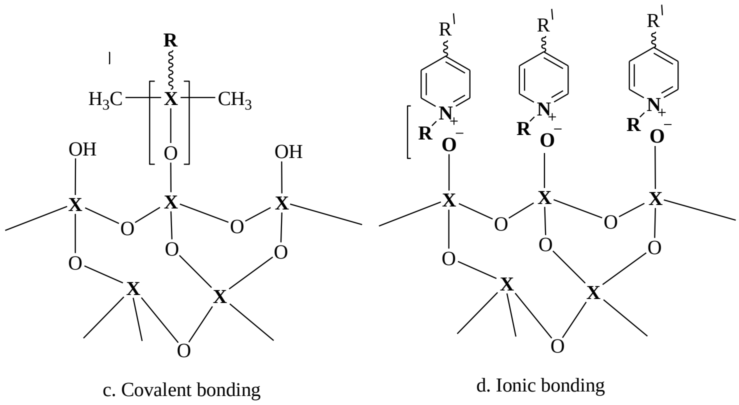

Figure 1. Classification of hybrid material: a and b are example of *class I* hybrid, c and d are example of *class II* (modified from Kickelbick (2007)).

2.1.1 Components of EOR hybrid commonly employed in CEOR
The CEOR hybrid's components mainly contain surfactants, polymers and nanomaterials in different molecular ratios depending upon the types of hybrid needed, and the nature of physicochemical properties required [18].

2.1.1.1 Surfactants
Surfactants are compounds capable of orienting themselves between the media and reduce the interfacial or surface tension between the displacing agents and oil phases [19]. Surfactants are made up of two different components; one component known as a *hydrophilic,* which has the ability to dissolve polar compounds while the other component, *hydrophobic*, dissolves non-polar compounds (figure 2). Possession of these unique properties qualifies surfactants for applications at different stages of petroleum production and processing operations [20]. Advanced technology usually combines surfactants and gases to generate foams, which is a potential chemical flooding agent for EOR applications [21]. From the literature reports, the application of surfactants in reducing the interfacial tension (IFT) between the different phases cannot be overemphasized. However, reducing IFT alone may not give the best oil recovery value, and hence, there is a need to focus on the manipulation of the overall oil sweep towards producer well.

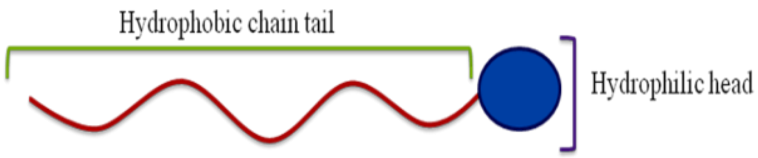

Figure 2. Schematic diagram showing the two components of surfactant.

The main challenge of injecting surfactants alone involves high economic loss, because at relatively higher temperatures their molecules are destroyed, precipitated and get deposited on the rock surface. They are also intolerant against shear stress between the rock pores, and sometimes finger into the oil bank due to less viscosity [22]. These factors affect the overall performance of surfactants which eventually lead to the recovery of the minimal amount of trapped oil.

2.1.1.2 Polymers
Polymers are used purposely to improve the rheological properties of the injected fluids (fluids containing water) by enhancing the viscosity so that the modified fluids would have the mobility less

than that of oil. As a result, more oil would be pushed towards the producing wells [23]. Water soluble polymers are the most commonly employed polymers for EOR applications. Reasons being that, their physicochemical properties have been investigated and have direct relationships with the ability to increase oil production in many tested oilfields. However, despite this improvement in enhanced oil recovery, several problems associated with polymer flooding were encountered, and led to poor efficiency over time [21]. These include, but are not limited to:

a. Mechanical and thermal instability
b. Effect of high salinity
c. Change in pH
d. Cross link behaviour
e. High molecular polymers plug low permeability reservoir rocks

2.1.1.3 Nanomaterials (nanoparticles)

Researchers within the field of EOR are increasingly investigating the effects of nano-fluids for their potential applications, not only in EOR but also in pharmaceuticals, cosmetics, electronics, material and living matter [24]. The idea of employing nanotechnology in EOR fields is to economically increase oil production and delays the oil field abandonment [25-26]. For successful EOR nanotechnology, nanoparticles formulations must meet the specific requirements like penetrating into the different rock-pore sizes at relatively low concentrations to alter wettability [27]. From our literature survey, we found out that several laboratory experiments and oil fields pilot applications of nanoparticles have been documented, and their potential effects to modify rock wettability and enhanced oil recovery were reported [28-32]. The mechanisms responsible for improving oil recovery using nanoparticles have been fully understood and are no longer controversial, as nanoparticles have the ability to change rock wettability from oil-wet to water-wet conditions [33]. In addition, Dang et al [34] also described the wettability modification near water-wet conditions as the reliable mechanism to understand the chemistry of formation rock surfaces. However, the utilization of nanoparticles to develop composite hybrids is still under scientific investigations. Efforts have been made to understand the synergistic relationship between composite materials containing nanoparticles to correlate with the improved oil recovery.

3. Literature review

This section presents the recent experimental studies from the literature, and highlights some advancements, advantages, and improvements in CEOR as a result of employing hybrid materials in EOR as compared with the conventional single system flooding techniques.

3.1 Surfactant-polymer hybrid

Shedid [35] investigated the feasibility of combining the surfactant (EZEFLO F75N) with Hydrolyzed Polyacrylamide polymer (HPAM-J120) in the low permeability carbonate reservoir (>10 md) with respect to IFT, viscosity and oil recovery rate. The result indicated that apart from lowering the IFT and improving the viscosity, the surfactant-polymer slug had an additional recovery factor (10 %) compared to the injection of the single polymer slug (7 %).

Sun et al [36] studied and compared the thermal resistance and oil recovery rate of foams that were generated from surfactant (mixture of sodium dodecyl sulphate + Imidazoline) and surfactant-polymer (mixture of sodium dodecyl sulphate + Imidazoline + Xanthan polymer). After aging at 90^{o}C, the hybrid slug of surfactant-polymer foam had four times the half-life drainage than the foam obtained from the mixture of surfactants alone. Furthermore, the oil recovery of surfactant-polymer foam was found to be twice that of surfactant foam, in core flood experiments at the same temperature.

Muhammed et al [37] had established that a surfactant-polymer hybrid increased hydrocarbon recovery compared to a single slug. Their studies revealed that the incorporating surfactant in the polymer gel solution could mitigate the effect of polymer plugging.

3.2 Polymer-nanomaterial hybrid

Zhu et al [12] developed polyacryamide (HAHPAM)/silica nanoparticles hybrid material to improve thermal stability, shear resistance, and salt tolerance behaviour of partially hydrolyzed polyacramide polymer (HPAM) for successful CEOR application. After comparing the rheological and enhanced oil recovery properties of hybrid and pure polymer at extreme reservoir conditions (temperature 85°C and 32,868 mgL-1 brine solution), they found that hybrid material of HPAM/silica nanoparticles had a better recovery factor, in addition to improved long thermal and shear stabilities as well as salt tolerance as compared to HPAM polymer alone. They concluded that the chemical interaction between silica nanoparticles and amide functional group in HPAM led to improved properties.

Similarly, Pu et al [38] synthesized a water soluble novel core-shell polymer, known as hyperbranched polymer (HBAMs) hybrid (figure 3), which is made up of polyamidoamide (subshell) and silica nanoparticles (core) via in-situ free radical polymerization reaction. Their investigations revealed that, this hybrid core-shell material could be a good CEOR agent due its unique thermal, shear degradation and salt tolerance behaviour.

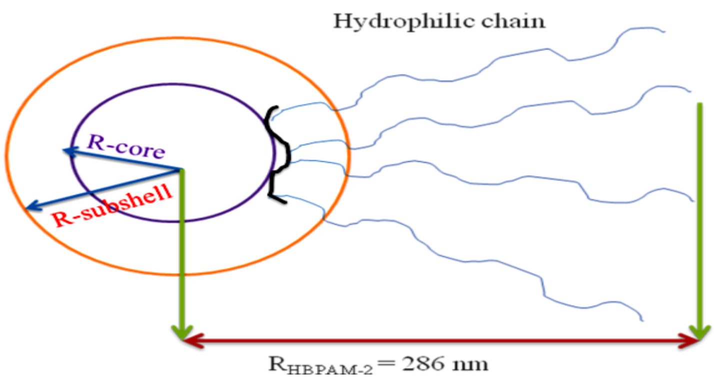

Figure 3. Schematic diagram of co-shell hybrid (modified from [38].

The review of ShamsiJazeyi et al [39] presented the applications of polymer-nanoparticles for enhanced oil recovery.

3.3 Surfactant-nanomaterial hybrid

Eftekhari et al [40] investigated the stability of nitrogen foam using nano-ash particles with alpha olefin sulfonate as a surfactant. They observed that the nano-ash particles, which are readily available as the waste product from a coal power plant, had the ability to stabilize the foam in the presence and absence of oil. Their research confirms that the nano-ash material is compatible with alpha olefin sulfonate surfactant (AOS), which could find other applications in the field of enhanced oil recovery nanotechnology.

Suleimanov et al [41] experimentally studied the effect of non-ferrous metal nanoparticles in a non-ionic surfactant solution. They discovered that the dispersion of metal nanoparticles in the surfactant solution significantly enhanced the reduction of surface tension in the oil media by nearly 90 %. And hence, recommended the use of nano fluids for application in enhanced oil recovery processes.

Worthen et al [42] revealed that silica nanoparticles could be a good composite material to obtain stable and viscous CO2 foam from caprylamidopropyl betaine (amphoteric surfactant). In their studies, they noticed that the interaction of a hydrophilic head group of the silica nanoparticles and the surfactant increased the contact angle between water, surfactant and CO2.

3.4 Surfactant-polymer-nanomaterials hybrid

El-hoshoudy et al [43] developed and successfully characterized a novel nano-composite of polyacrylamide-SiO_2 using surmers as a functional surfactant. After investigating the rheological properties of the novel nano-composite with respect to salinity, shear rate, temperature, and concentration, they observed that the novel composite showed non-Newtonian behaviour and improved properties (thermal, shear and salinity). In the core flood experiment, the novel composite recovered up to 60 % of the residual oil saturation.

Sharma et al [44] investigated the synergistic effects between polymer, surfactant and nanoparticles (SPN) in a single hybrid system to find out the justification of additional oil recovery after comparing with the surfactant-polymer system (SP). The result revealed that significant improvement of properties like viscosity, IFT and pressure drop were noticed in hybrid material (SPN). Similarly, core flood experiments were conducted by flooding with an SPN hybrid at high temperature and high pressure conditions. More than 60 % oil recovery was recorded, which could be due to capillary end effect, as compared to SP flooding.

Nguyen et al [45] synthesized surfactant/polymer/SiO_2 inorganic composite for application in EOR processes under extreme reservoir conditions. They recommended that this composite material could find suitable applications in EOR by increasing the oil displacement efficiency.

4. Limitation of CEOR hybrid materials

Despite the increasing research in finding the promising hybrid composites for successful applications in EOR, sudden changes in reservoir conditions (temperature, pressure, salinity) and crude properties are the main challenges which affect the success of CEOR hybrid flooding for improving oil recovery [46]. The compatibility between the hybrid components are strongly affected by a high temperature, as well as the concentrations of ions in brine solution. Petroleum engineers and scientists are still putting efforts to understand the mechanism of compatibility with respect to physical and chemical conditions in porous media. Apart from this, full field operational implementation of designed projects using hybrids CEOR is still facing economic and technical challenges due to the high cost of chemicals, and finally lack of good engineering technology for successful execution of the projects [47].

5. Conclusion

It is concluded that the synergistic actions between the hybrid components are the main reason that CEOR hybrids are finding wide applications in the field of EOR. This area of oil recovery requires extensive research, particularly on the compatibility studies and the overall performance of CEOR hybrids with respect to changes in physicochemical conditions of reservoirs, as well as changes in crude properties. This would help exploration Engineers to understand and implement successful EOR technologies.

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