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Review Article

Application of Nanofluids and Nanocomposites for Enhanced Oil Recovery

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Abstract *- The low recovery of oil (only one-third) is mainly related to the displacement efficiency of porous media, which is influenced by wettability and interfacial tension. Since a large amount of oil deposits, two third of the original oil-in-place is trapped by the capillary forces, and there is a need to recover residual oil by improving oil recovery techniques. Although gas, thermal, microbial, and chemical injection is very popular and highly used techniques, they have some disadvantages. Therefore, tertiary oil recovery techniques, such as the application of nanofluids and nanocomposites, may solve this problem. The selection of appropriate techniques depends on the reservoir and economics. The mobility ratio and the mechanisms for nano-enhanced oil recovery have also been explained. Silica, zinc oxide, titanium dioxide, carbon-based nanoparticles, graphene quantum dots, graphene oxide nanosheets, and anionic surfactants are widely used in enhanced oil recovery research. Nanocomposites were discussed recently prepared, including potassium chloride/silicon dioxide/xanthan and zinc oxide/silicon dioxide/xanthan nanocomposite and others. The reviewed literature experimental data has shown that it is possible to increase the enhanced oil recovery in the 10 to 79% range depending on the applied nanofluid or nanocomposite.*

Keywords *- Nanofluids, Nanocomposites, Enhanced oil recovery, Wettability, Interfacial tension.*

1. Introduction

The life of today's man is unimaginable without the daily use of energy, such as electricity, fuel or other types of energy. Energy demand is growing across many countries in the world as people get richer and populations increase [1]. Total energy consumption in the world is growing by 2% every year [2]. Thus, the ever-increasing need for energy has imposed a demand for new energy sources. However, although we are witnessing a major shift towards developing and using renewable energy sources, the oil and gas industry remains the world's most important fuel source.

Renewable energy techniques are unlikely to meet the demands effectively because they are costly and insufficient for energy demand satisfaction [3]. The low recovery of oil is mainly related to the displacement efficiency of porous media, which is influenced by wettability and interfacial tension (IFT)[4]. Therefore, there is a need to improve oil recovery techniques since a large amount of oil deposits still remain after conventional recovery methods.

Enhanced oil recovery (EOR) is the process of extracting additional crude oil from oil reservoirs. Hydrocarbon is the primary energy source for humankind and is believed to remain so in the near future. The crude oil extraction process can be classified as primary, secondary, and tertiary oil recovery stages. Usually, through the primary (natural energy-driven) and secondary (water-driven) stages, 40-50% of crude oil can be recovered [5]. It is estimated that only one-third of the oil can be extracted by conventional production methods [6]. That is, two third of the original oil in place (OOIP) is trapped by the capillary forces.

For this reason, it is important to use enhanced oil recovery methods to recover residual oil [7]. Therefore, the tertiary oil recovery techniques (nanofluids) are crucial to extract further residual crude oil trapped within reservoir pores and channels. They can be classified into chemical flooding methods, thermal recovery methods, and miscible flooding methods (gas injection). The selection of techniques is completely dependent on the reservoir and economics.

Enhanced oil recovery (EOR) is the process of extracting additional crude oil from oil reservoirs. EOR methods are classified as chemical, thermal, and pressure maintenance ones. Among these methods, the application of the chemically enhanced oil recovery (CEOR) method has been favoured due to its higher efficiency, lower capital costs and techno-economic feasibility [8]. Different chemical methods can alter mobility control, the interaction between fluid to fluid, and fluid to rock surface and improve oil recovery.

The main chemical EOR methods are based on the application of polymers and surfactants (Fig.1). The principle of work of the *polymer-based EOR method* is based on using a polymer to modify the shear viscosity [9] of the injected fluid and hence influence on mobility ratio [10]. The most common polymer used is partially hydrolysed polyacrylamide (HPAM) due to its cost, availability, and easy handling [11].

According to a business wire report [12], the global market for chemically enhanced oil recovery, estimated at US$776 Million in the year 2020, is projected to reach a revised size of US$1.14 Billion by 2027, growing at a CAGR of 4.9% over the period 2020-2027. Polymer flooding is projected to record a 5.3% CAGR and reach US$593 Million by the end of 2027.

The principle of work of the *surfactant-based EOR methods* is based on using surfactants to modify the wettability of porous media by controlling the interfacial tension (IFT) between the production and injection fluids. By injecting surfactants, alcohol and polymers, the oil is successfully displaced [13]. By applying primary oil recovery methods, up to 20% of the OOIP can be extracted, while by applying EOR, additional amounts of crude oil from oil deposits increase by about 30-60% [14]. However, approximately 50% of oil in the reservoirs cannot be recovered by conventional chemical flooding [15]. Miscible flooding is a technique in which the displacing phase (CO_2, flue gas, liquefied petroleum gas, methane, etc.) mixed up with crude oil enhanced oil recovery thanks to the disappearance of interfaces. CO_2-enhanced oil recovery helps to reduce global greenhouse gas emissions by sequestering CO_2 in subterranean geological formations [16]. Gas injection offers advantages, such as reduction of oil viscosity and added pressure in the reservoir for oil production.

According to the International Energy Agency (IEA), the systematic application of all available EOR technologies could unlock approximately 300 billion barrels of recoverable oil [17]. The enhanced oil recovery market is expected to register a CAGR of more than 2% during the forecast period 2022-2027 [17]. The techniques of gas injection, thermal injection and microbial injection are the most represented; herein will be discussed out the chemical injection.

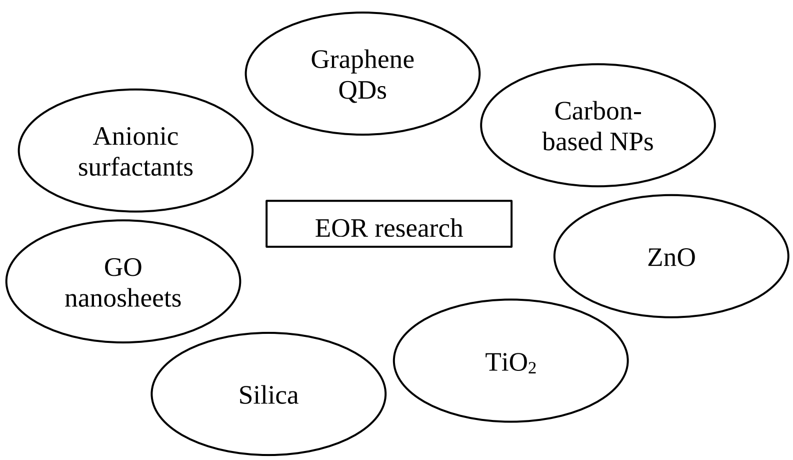

Fig. 1 The main chemical EOR methods based on the application of anionic surfactants, graphene QDs, carbon–based nanoparticles (NPs), metal oxide NPs, silica and graphene oxide (GO) nanosheets

The oil recovery by water, surfactant [63], or gas injection exhibited only a low recovery, less than 10% of OOIP. Oil recovery during the injection of regenerated foam was improved significantly, with up to 78% of OOIP produced [19].

The thermal recovery methods use high temperatures to decrease highly viscous oil viscosity. These methods include steam flooding, huff and puff, and fire reservoir technologies [20].

Although chemical EOR methods are promising for improving crude oil extraction, these methods can have some disadvantages due to the large amount of chemicals required and the high cost, as well as their decomposition in the reservoirs. In addition, they have negative environmental consequences, such as impact on groundwater, which is an obstacle to their widespread application.

Therefore, the solution to this problem can be offered by nanotechnology. The application of nanotechnology for improving oil recovery is based on the dispersion of nanomaterials (nanoparticles, nanosensors, nanocomposites, nanofluids) [21-22] into specific fluids such as water, ethanol, or other dispersants to form nanofluids. Nanofluids have proven excellent for removing trapped oil from reservoirs by increasing wettability, controlling mobility, reducing interfacial tension (IFT) and improving rheology [23].

This paper will discuss the application of nanocomposites and nanofluids for EOR. In Fig. 4, the number of research papers published per year in the last ten years for searched keywords “nanocomposites for enhanced oil recovery” and “nanofluids for enhanced oil recovery” on ScienceDirect. From Fig. 2, one can see that a higher number of research papers are present with nanocomposites than with nanofluids, but these results should be considered with caution because many nanocomposites end up being used dispersed in a liquid and are again called nanofluids.

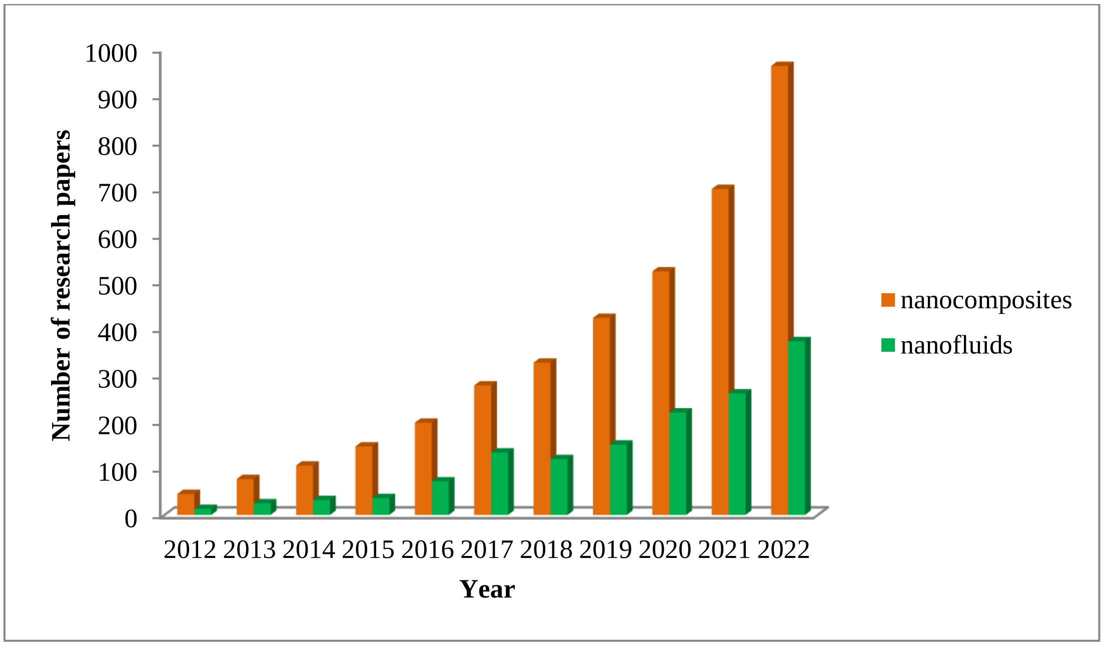

Fig. 2 Number of research papers published per year in the last ten years for searched keywords “nanocomposites for enhanced oil recovery” and “nanofluids for enhanced oil recovery” on ScienceDirect

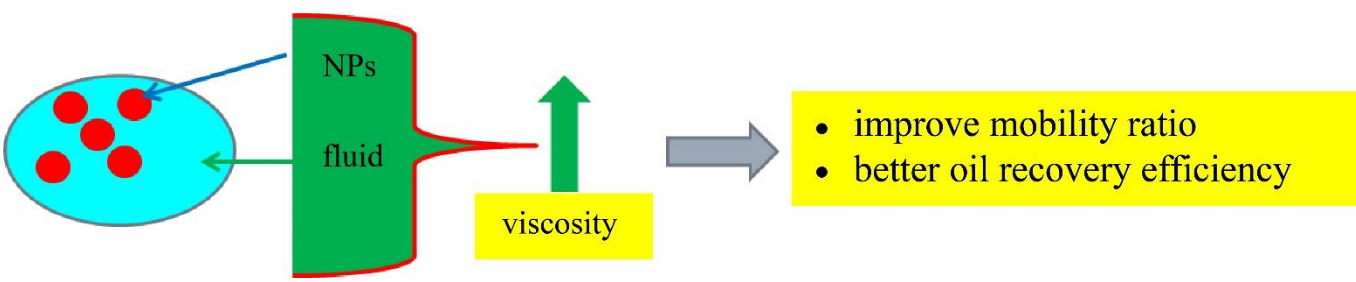

Fig. 3 Improvement of the mobility ratio

1.1. The Mobility Ratio

The mobility ratio is one of the most influential factors that deteriorate oil recovery by water flooding because water cannot completely sweep the oil due to its lower viscosity [24-25]. A polymer solution is made by mixing polymers with water, boosting the apparent viscosity of the displacing fluid to lower the mobility ratio. The mobility ratio (M) is the ratio of the mobility of displacing fluid to the mobility of the displaced fluid [26] (Eq.1).

$$M = \frac{Mobility\ of\ displacing\ fluid}{Mobility\ of\ displaced\ fluid} \quad (1)$$

If M is higher than 1 (M >1), this indicates that water is more mobile than oil and would pass through the oil zone, creating an early breakthrough and therefore resulting in low displacement efficiency. However, where the mobility ratio is less than or equal to 1 (M ≤ 1), this is considered to be a favourable state for oil displacement. Lack of a good mobility ratio between the oil and injection fluid often results in viscous fingering, leading to inappropriate sweep efficiency because the oil viscosity is considerably greater than that of the injection fluid. Therefore, it is paramount to control the oil/water mobility to improve the sweep efficiency to achieve higher oil recovery. The idea of adding NPs into the fluid has been proposed to increase the viscosity up to the optimum level, with the net effect resulting in an improvement of the mobility ratio and hence better oil recovery efficiency (Fig. 3) [3].

1.2. The Mechanisms for Nano-Enhanced Oil Recovery

Many mechanisms behind increasing oil recovery induce alteration of surface wettability toward more water-wet when displacing oil by nanofluids. The responsible mechanisms for nano-enhanced oil recovery by application of nanofluids are attributed to an increment in the viscosity of injection fluid, decrement in oil viscosity, reduction in interfacial and surface tension, and alteration of wettability in the rock formation [27]. The EOR mechanisms of nanofluids are based on the adsorption of nanoparticles at the oil/water interface that reduces the interfacial tension [28], wettability alteration [29-30], and increased structural disjoining pressure [31]. By decreasing the interfacial tension, the recovery rate increases [32]. A recently developed nanofluid based on modified carbon black (MCB) nanoparticles was recently developed for EOR in low permeability reservoirs [33].

Final oil recovery was significantly increased by 27.27% with spherically dispersed (MCB) nanoparticles than with carbon black (CB) nanoparticles and surfactants. MCB nanoparticles with an average size of 72.3 nm reduce the oil–water interfacial tension (IFT) to 10^{-2} mN/m and change the surface wettability of sand particles. The EOR mechanisms of the MCB nanoparticles were explained by its adsorption on the surfaces of sand particles and forming larger aggregates that bridge across pores or throats which improved swept volume and its synergistic effects with oil displacement efficiency.

Due to the low porosity and low permeability in unconventional reservoirs, a large amount of crude oil is trapped in micro-to nano-sized pores and throats, which leads to low oil recovery.

Wang *et al.* (2022)[34] indicated that the nanofluid at the oil/water/solid three-phase interface reduces the oil/water interfacial tension, and the three-phase contact angle dropped from 135° (oil-wet) to 48° (water-wet). The EOR mechanisms of nanofluids in low permeability reservoirs are based on the formation of the high-strength interface film by the nanofluid, inhibiting the coalescence of oil droplets and improving the flowing ability [34]. More than 2/3 of the total hydrocarbon reserves in China are low permeability reservoirs and have become the strategic substitute for conventional energy sources [64]. These reservoirs contain micro and nano-pores with pore sizes ranging from 0.01 to 10 µm [36]. The development of low permeability reservoirs is limited due to the small pore sizes, which increase capillary force and lead to high flow resistance and poor injectivity [37].

2. Nanofluids in EOR

Even though various nanoparticles (NPs) were used and discovered to play imperative roles in EOR, they have not been widely used in oilfield development. NPs tend to agglomerate due to their large surface area and surface activity. Using surfactants is a significant strategy to improve the stability of NPs in fluids [38]. Moreover, the presence of nanoparticles increases the effect of surfactant solution on oil recovery processes by changing the IFT value of the surfactant/oil interface more effectively and rheological properties [39]. NPs are usually injected in the form of nanofluids. Nanofluids are fluids containing NPs (dispersed solid phase) with less than 100 nm of average size in the colloidal suspension (liquid phase). Solid particles conduct heat much better than liquids. Nanofluids are also called smart fluids. Nanofluids can reduce the viscosity of extra-heavy oil [40]. Silica NPs [41] and carbon-based NPs have been used in most EOR experiments.

Nasr *et al.*, 2021[23] reported the EOR performances of nitrogen-doped graphene quantum dots (N-GQDs) with different GQD-sizes and nanofluid concentrations. They found that 0.5 mg mL^{-1} of N-GQDs nanofluids altered the wettability of carbonate rocks and reduced the hydrocarbons/water IFT in mN/m unit ($IFT_{oil/water}$ from 16.88 to 0.975 and $IFT_{n\text{-}heptane/water}$ from 50.24 to 27.87, respectively). Compared to water flooding, this nanofluid increased oil recovery factors by 18% and 14% from carbonate and sandstone core samples.

Silica nanoparticles (SiNPs) are desirable to use as they are environmentally friendly, cheap, can be easily controlled by surface modification and are natural sandstone composition. Youssif *et al.* (2018)[42] found that the ultimate recovery of initial oil in place increased by 13.28% with 0.1 wt% of SiNPs of 20 nm in size in sandstone cores in flooding experiments.

Hassan *et al.* (2022)[38] found increasing in stability and viscosity of zinc oxide (ZnO) and silicon dioxide (SiO_2) NPs by 37 and 43 % when the composite nanofluids ZnO/SiO_2 was used, while upon introducing polyvinylpyrrolidone (PVP) and sodium dodecylbenzene sulfonate (SDBS) (nano-surfactants) that increased 54 and 44 %, respectively.

Shirazi *et al.* (2019) found that applying smart water with TiO_2 nanoparticles in carbonate reservoirs changes wettability, reduces IFT, and improves oil recovery. Carbonate reservoirs contain fractured networks and oil/mixed-wet matrices. Due to this complex structure, the amount of oil recovery via primary and secondary methods is about 20% to 40% of the original oil in place [43].

Suleimanov *et al.*, 2011 [32] studied nanofluids based on an aqueous solution of anionic surfactants with the addition of light non-ferrous metal nanoparticles for enhanced oil recovery. The nanofluid reduces surface tension by 70–90% on an oil boundary compared to aqueous surfactant solution. EOR has been significantly increased by using nano-suspension. The dimensions of nanoparticles determine the main feature and properties of nanofluid. When NPs are suspended, they can increase the sedimentation stability and thermal, optical, stress–strain, electrical, rheological and magnetic properties [32].

The application of nanofluids for EOR in low saline environments is confined due to its instability. Khoramian *et al.*, 2022[44] developed a novel nanofluid based on anionic surfactant and graphene oxide nanosheets (GONs) that preserved a high salinity of 6 wt% sodium chloride (NaCl) than bare silica nanofluid. Adding NaCl significantly improved the viscosity of Janus-GONs fluid, which was 1.48 cP compared with 0.07 cP for silica fluid. Janus-GONs yielded a considerable oil recovery of 79 %, compared to 53 % for silica.

A novel nanofluid based on the sulfonated graphene (*G-DS-Su*) at concentrations of 0.5 and 2 mg/mL could EOR for 16 and 19% [45]. These nanofluids tend to form emulsions. The wettability alteration may be due to both forming a wedge film in the oil/nanofluid/solid three-phase contact region and the adsorption of *G-DS-Su* onto the sandstone due to the π-π and n-π interactions between the functional groups of sand and *G-DS-Su*.

Surface-active ionic liquids (SAILs), i.e. 1-dodecyl-3-methylimidazolium chloride, [C_{12}mim]Cl in combination with Al_2O_3 nanoparticles, have been successfully applied to increase oil recovery [46]. The presence of 0.05 wt% Al_2O_3 reduced the adsorption of the surfactant-polymer formulation [C_{12}mim]Cl and 1.0 wt% PVP on carbonate rocks in brine (5.0 wt% NaCl). The surfactant-polymer formulation allows additional oil recovery of 10.4 % OOIP in comparison with 14.8 % OOIP obtained with the nanofluid [46].

Zhao *et al.* (2022) [47] studied the interaction between naphthenic arylsulfonate surfactant (NAS) and petroleum components at low surfactant concentrations. They found that micellar solubilization plays a significant role during the surfactant flooding for the EOR process. The system NAS/alkanes show the reduction of IFT value (2.53, 1.37 and 0.36 mN,m^{-1}) with the increase of NAS concentration of 0.1 wt%, 0.2 wt% and 0.4 wt%, respectively. They attributed these findings to the formation of interfacial membranes from the aggregation of amphiphilic molecules at the oil-water interface, which enhances the surface pressure, leading to decreased interfacial energy and IFT [48]. Polymeric nanoparticle (NP) suspensions of SiO_2, ZnO, and TiO_2 nanoparticles were used as carrier fluids for CO_2 - loaded (carbonated) water injection and used as carbonated fluids for enhanced oil recovery from sand packs [49]. The CO_2 absorption has been improved by nanoparticles in the base fluid, while the addition of salt (3 wt% NaCl) induced aggregation of NPs in suspensions and reduced their CO_2 loading capacity. Flooding experiments showed that the carbonated polymeric NP suspensions are able to increase oil recovery even at a higher temperature and salinity (an additional 12–20% of OOIP) than their non-carbonated peers.

A novel polymeric nanofluid *Cissus populnea* nanoparticle (CPNF) has been successfully synthesized [65], and rheological properties were compared with *Cissus populnea* (CP) solution and commercial polymer xanthan. The research results showed that increasing the temperature of the CP and CPNF solution is accompanied by an increase in viscosity, while the opposite happens with xanthan, i.e. with an increase in temperature, the viscosity decreases. Furthermore, CPNF decreases IFT value and increases oil recovery by 26%. The evaluation of energy consumption shows that it is a more cost-effective application of polymeric nanofluid than conventional EOR chemicals.

An innovative polyoxyethylated graphene oxide-based nanofluid (P-GO-O) for enhanced oil recovery was prepared [51]. The P-GO-O nanofluid was capable of changing the oil-wet surface to the water-wet surface. The P-GO-O nanofluid (17.2%) has a higher oil recovery ratio than the GO-O (octadecylaminated graphene oxide) nanofluid (6.7%). According to [51], amphiphilic carbon-based nanomaterials with a Janus behavior have attracted much attention in chemical-enhanced oil recovery.

Rashidi *et al.*, 2021[52] studied the effect of calcium carbonate nanofluid on the wettability alteration of sandstone rock samples was studied. The average particle size of nanoparticles was 55.4 nm. The rock samples were aged in oil to ensure desired oil-wet conditions. The 2-D oil-wet glass micromodel results showed an alteration of wettability from oil-wet to water-wet for all nanofluid concentrations. The wettability alteration was achieved at higher temperatures (80 °C) than at room temperature (25 °C). The contact angle was changed with the optimal nanofluid concentration (0.025 wt%) to 46.99° at 25 °C and 28.7° at 80°C from 116.41° non-treated surface. An additional ultimate oil recovery factor was 20% with the optimal nanofluid concentration than normal water flooding (Rashidi *et al.*, 2021)[52].

3. Nanocomposites in EOR

The NPs can be applied in different manners for EOR. The nanoparticles can be dispersed in brine, surfactant, polymer and polymer-coated nanocomposite [53]. Zargar *et al.*, 2020 [54] prepared a green nanocomposite (NC) for EOR applications. The titanium oxide NPs were prepared by the green method from the *euphoria condylocarpa* extract and grafted on the surface of the quartz. This nanocomposite NC were dispersed in desilted water, seawater and low salinity water (seawater dilution) to prepare the novel nanofluids for reducing the interfacial tension (IFT) and contact angle between crude oil and water on the surface of carbonate rocks. An additional oil recovery of 21% OOIP was achieved when 1000 ppm of TiO_2/Quartz-nanofluid (DWN1000) was dispersed in distilled water due to the reduction in IFT from 36.4 to 3.5 mN/m and improving the wettability alteration towards a stronger water-wet system from 103° to 48° contact angle (Zargar *et al.*, 2020) [54].

In the research [55], the nanocomposite chloride/silicon dioxide/ xanthan (KCl/SiO_2/Xanthan) NCs were prepared and used for EOR. They used 1000 ppm of NC and improved the oil recovery for 17.05% OOIP due to altering the wettability of rocks from oil-wet to water-wet, reducing the IFT and contact angle.

Another example of green-synthesized oxide/silicon dioxide/xanthan (ZnO/SiO_2/xanthan) nanocomposite has been prepared [56]. They were able to create better interactions between crude oil–polymer–nanoparticles–carbonate rocks resulting in a big improvement in oil recovery (19.3%), a huge reduction in IFT (93%), and significant alteration in wettability towards a water-wet system.

The research carried out by Ju and Fan, 2009[57] showed that using polymer coated-SiO_2 nanocomposite was able to increase oil recovery up to 21% due to the alteration of wettability while the effect of IFT reduction on the residual oil was not significant.

On the contrary, silica NPs coated by polymer were not able to change the IFT and wettability significantly but could increase residual oil recovery by about 10% [58]. In research carried out by Zhang *et al.*, 2016 [59], silicon was dispersed in polyethylene glycol, and the oil recovery was up to 17%, achieved by the combined effect of IFT reduction and wettability alteration. Rezvani et al. 2018 [60] stated that using a fabricated Fe_3O_4/chitosan nanocomposite successfully produced an extra 10.8% OOIP due to modifying the IFT and wettability behaviours.

Chen *et al.* (2021) [61] prepared temperature-sensitive iron oxide nanoparticles (TSIO) and used them in an enhanced oil recovery process. Iron oxide (IO) was modified in situ with poly (N-isopropylacrylamide) (PNIPAM) and styrene sulfonate groups.

They showed that by increasing the temperature, TSIO greatly reduced the IFT of the oil and water and the wettability of the rocks improving the oil recovery. At 50 °C, the recovery rate raised to 84.02%, while at ambient temperature increased from 65.78% to 74.15%.

Thermostable and highly water-soluble polymers are essential for polymer flooding and one of the most effective methods used in enhanced oil recovery (EOR) in high-temperature (HT) offshore reservoirs. The nanocomposites (GO–P(AM-NVP) were prepared from copolymers P(AM-NVP) obtained from acrylamide (AM) and N-vinylpyrrolidone (NVP) monomers and then covalently coupled with graphene oxide (GO). The thermal and chemical stabilities of the brine-dispersed P(AM-NVP) copolymers annealed at 123 °C (the WT Miocene reservoir temperature) and the GO–P(AM-NVP) nanocomposite dispersion annealed at 135 °C (the WT Oligocene reservoir temperature) for 31 days were observed through the visual inspection and viscosity testing. Results indicated that the dispersions of the P(AM-NVP) copolymers and P(AM-NVP) copolymers conjugated on the GO nanosheets exhibited excellent thermal and chemical stabilities [62].

4. Conclusion

Silicon dioxide, zinc oxide, titanium dioxide, carbon-based nanoparticles, graphene quantum dots, graphene oxide nanosheets, and anionic surfactants are widely used in enhanced oil recovery (EOR) research. The green methods were used to prepare titanium oxide nanoparticles from the *euphoria condylocarpa* extract. From nanocomposites were discussed recently prepared potassium chloride/silicon dioxide/ xanthan (KCl/SiO_2/xanthan) and zinc oxide/silicon dioxide/xanthan (ZnO/SiO_2/xanthan) nanocomposite and thermostable and highly water-soluble polymer nanocomposites graphene oxide- copolymers P(AM-NVP), obtained from acrylamide (AM) and N-vinylpyrrolidone (NVP) monomers and then covalently coupled with graphene oxide (GO) (GO–P(AM-NVP).

Presented research results indicate that the most effective nanofluid is based on anionic surfactant and graphene oxide nanosheets (GONs) with the addition of NaCl with significant improvement of viscosity of Janus-GONs fluid and yields a considerable oil recovery of 79 %, compared to 53 % for silica. Then follow, the application of smart water with TiO_2 nanoparticles in carbonate reservoirs which recovered about 20% to 40% of the original oil in place via primary and secondary methods. A similar final oil recovery was achieved with modified carbon black nanoparticles and novel polymeric nanofluid *Cissus populnea* nanoparticles (CPNF) at 27.27% and 26%, respectively.

While calcium carbonate nanofluid achieved an additional ultimate oil recovery factor of 20%, carbonated polymeric NP suspensions can increase oil recovery for an additional 12–20% of original oil-in-place (OOIP) than their non-carbonated peers. A novel nanofluid based on the sulfonated graphene (*G-DS-Su*) at concentrations of 0.5 and 2 mg/mL was able to EOR for 16 and 19%. Nitrogen-doped graphene quantum dots increased oil recovery factors by 18% and 14% from carbonate and sandstone core samples. The polyoxyethylated graphene oxide-based nanofluid has shown an oil recovery ratio of 17.2%. The surfactant-polymer formulation, surface-active ionic liquids, i.e. 1-dodecyl-3-methylimidazolium chloride, $[C_{12}mim]Cl$ in combination with Al_2O_3 nanoparticles allow additional oil recovery of 10.4 % OOIP in comparison with 14.8 % OOIP obtained with the nanofluid.

All these results show that an additional amount of oil can be extracted using nanotechnology. It should particularly continue with the research and development of new nanofluids and nanocomposites for EOR application.

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