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Review

Nanotechnology in Enhanced Oil Recovery

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Abstract: Nanoparticles (NPs) are known as important nanomaterials for a broad range of commercial and research applications owing to their physical characteristics and properties. Currently, the demand for NPs for use in enhanced oil recovery (EOR) is very high. The use of NPs can drastically benefit EOR by changing the wettability of the rock, improving the mobility of the oil drop and decreasing the interfacial tension (IFT) between oil/water. This paper focuses on a review of the application of NPs in the flooding process, the effect of NPs on wettability and the IFT. The study also presents a review of several investigations about the most common NPs, their physical and mechanical properties and benefits in EOR.

Keywords: interfacial tension; NPs; nanotechnology; wettability; EOR; nanofluids

1. Introduction

Nanotechnology is defined as a new technology for making applicable matters, systems and devices using nanosized materials, as well as new phenomena and properties at the nanoscale (1–100 nanometers) [1]. Adding certain Nanoparticles (NPs) to injection solutions can significantly benefit enhanced oil recovery (EOR), with advantages such as wettability alternation, changes in fluid properties, improving the trapped oil mobility, enhancing the consolidation of sands and decreasing the interfacial tension (IFT) [2,3]. Nanomaterials are novel alternative methods to be applied as new EOR techniques and solve residual oil problems in heavy and semi-heavy oil reservoirs in last decade [4–8]. Figure 1 shows different EOR methods categorized into four major groups [9–11]. Nanoparticles and nanofluids, as interdisciplinary sciences, have great significance in the fields of thermal engineering, materials and nanoscience come across. Over the last decade, they have developed greatly and uncloaked their potential applications in EOR [12–16]. There are many studies that have indicated improved EOR processes by NPs and nanofluids in the past decade [5,7,17–19]. In several works, nano- and microspheres have been used for mobility control and have indicated excellent results in decreasing water cut, improving sweep efficiency and oil recovery [20–22]. In addition, micro and nanospheres can reduce the capillary force and water relative permeability and finally change the water flow path in porous media [22]. Moreover, NPs have suitable resistance to degradation at high salinity and temperature in oil and gas reservoirs. Some studies also investigated surfactant solutions with NPs as nanofluids for improving oil recovery in harsh reservoir conditions [21].

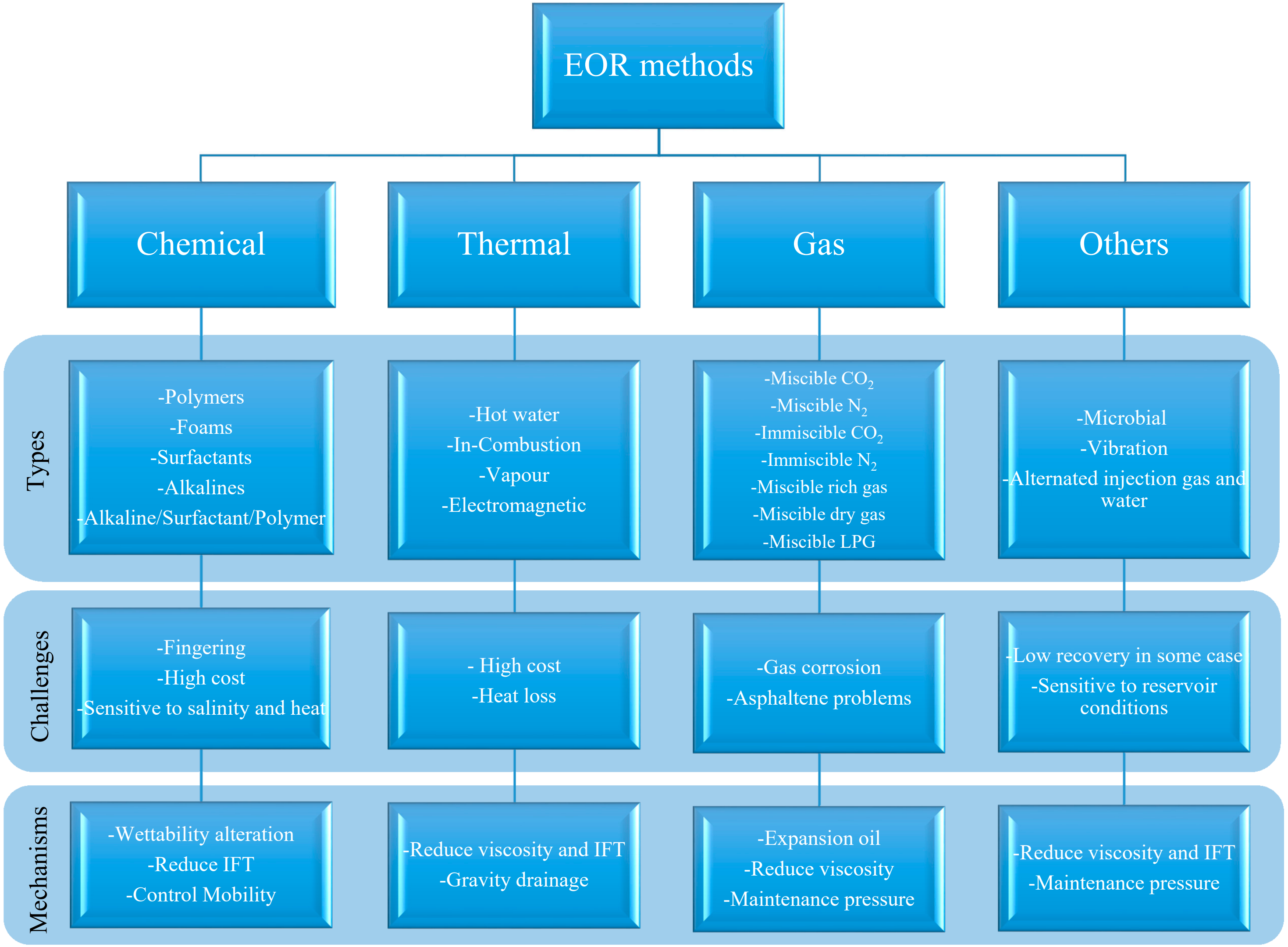

Figure 1. Flow sheet of enhanced oil recovery methods, mechanisms and challenges.

Many researchers have used nanomaterials to reduce the viscosity of bitumen and heavy and semi-heavy oil. Based on some experimental results, NP concentration, size and type are different parameters that affect the reduced viscosity mechanism of heavy oil [23–29]. In addition, much research has shown that wettability alteration and IFT reduction between fluids and rocks are two main mechanisms that NPs have great potential to enable on micro- and nanoscales (Figure 2) [30].

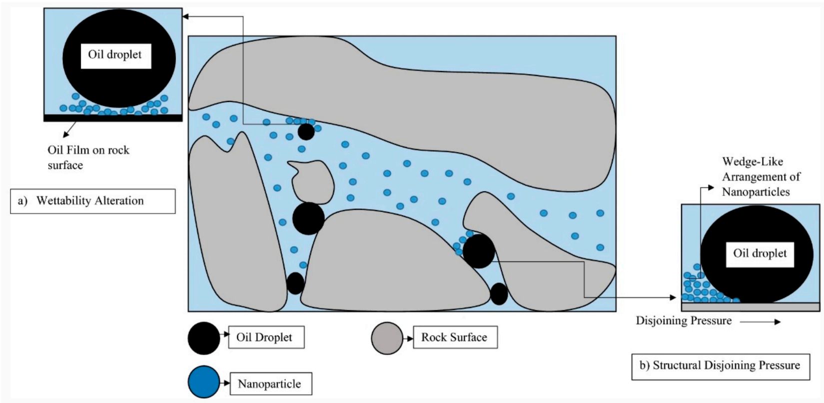

Figure 2. Mechanism of NPs EOR. (**a**) Changing wettability and (**b**) structural pressure by NPs [31].

To review the applications of NPs in EOR technology, more than 400 research papers on the issue or related to these subjects (2010–2020) were selected. Then, 100 up-to-date articles, with priority given to the latest research in recent years (2015–2020), were chosen for detailed evaluation. All of the statistics are extracted from the Web of Science, Scopus, Google Scholar and SCImago journal databases. According to these data, between 2007 and 2020, 498 documents were published. Figure 3 reports the number of scientific publications and document types relating to NPs for EOR.

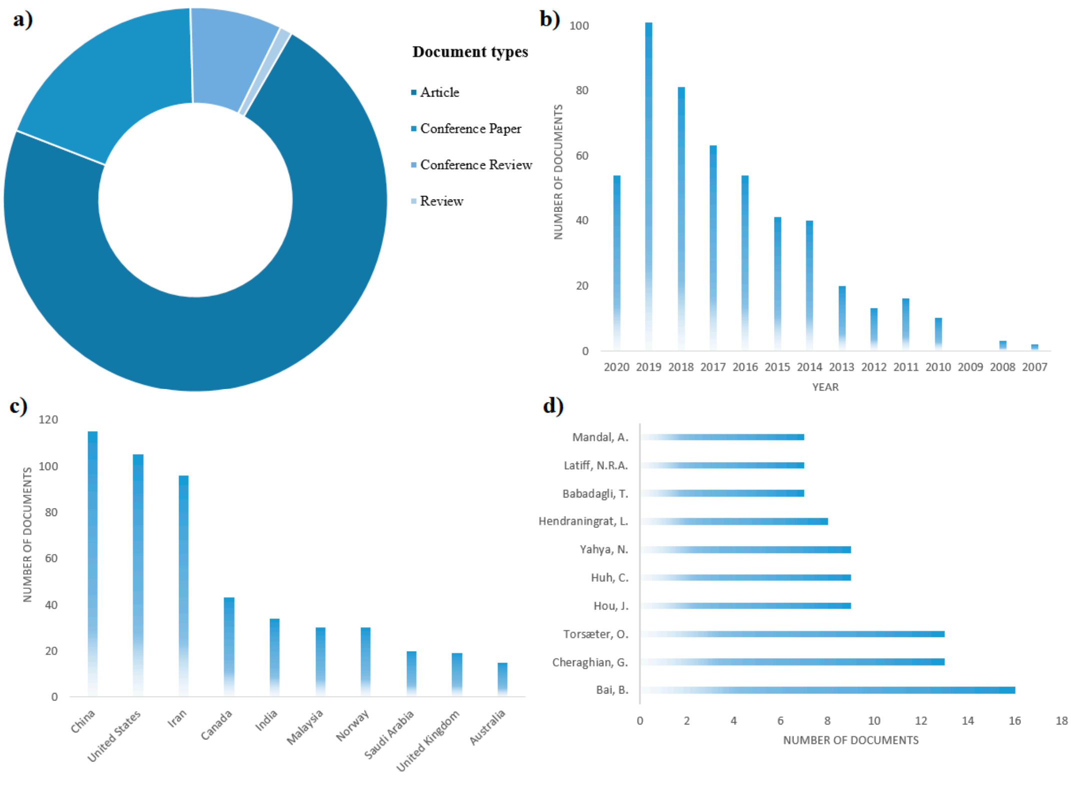

Figure 3. (**a**) Types of documents, (**b**) the number of documents per year, (**c**) the number of documents based on countries and (**d**) documents by author.

The present review assesses new nanoparticle investigations in EOR and assesses some of the recent nanomaterials applications in EOR technology. The goal of this study is to introduce an overview of nanotechnology in EOR and the physical properties and other characteristics of nanoparticles in EOR.

2. Effects of Nanoparticles on Oil Recovery

A combination of surfactants and NPs causes the release of oil drops, which are trapped in thin throats and microchannels of the reservoir rock. These phenomena are related to some agents that lead to an increase in oil recovery; these include parameters such as the wettability alteration of reservoir rocks, spontaneous emulsion formation, changing IFT between reservoir fluids and flow properties of porous media [32–35].

Over the years, various NPs were evaluated to improve oil recovery. For instance, Ehtesabi et al. [36] found that the TiO_2 NPs achieved an 80% increase in oil recovery in oil-wet sandstone. Shah [37] investigated the performance of oil recovery with CuO nanoparticles. The results showed that CuO nanoparticles lead to a 71% increase in oil recovery. Kanj et al. [38] studied the flooding performance of modified carbon nanoparticles. They found that carbon-based fluorescent NPs increase the oil recovery factor in carbonate reservoir by more than 96%.

In order to investigate the emulsion stability and polymer solutions in reservoir conditions, several NP-modified polymers have been developed by researchers. Wang et al. [39] focused on the effect of Na–montmorillonite (Na–Mt) particles and hydrolyzed polyacrylamide (HPAM) on emulsion stability. They also studied the interfacial properties of water–oil, dilatational viscoelasticity, IFT and zeta potentials. The results indicated that increasing NP concentrations cause a decrease in the IFT and zeta potential of oil drops and increases the dilatational viscoelasticity. With increasing NP concentrations, dilatational viscoelasticity and IFT were increased, while zeta potential became stable after 250 mg/L concentration. Actually, in the heavy oil reservoir, polymer-coated nanoparticles can change the pH conditions of the reservoir and cause recovery of heavy oil, which is shown in Figure 4.

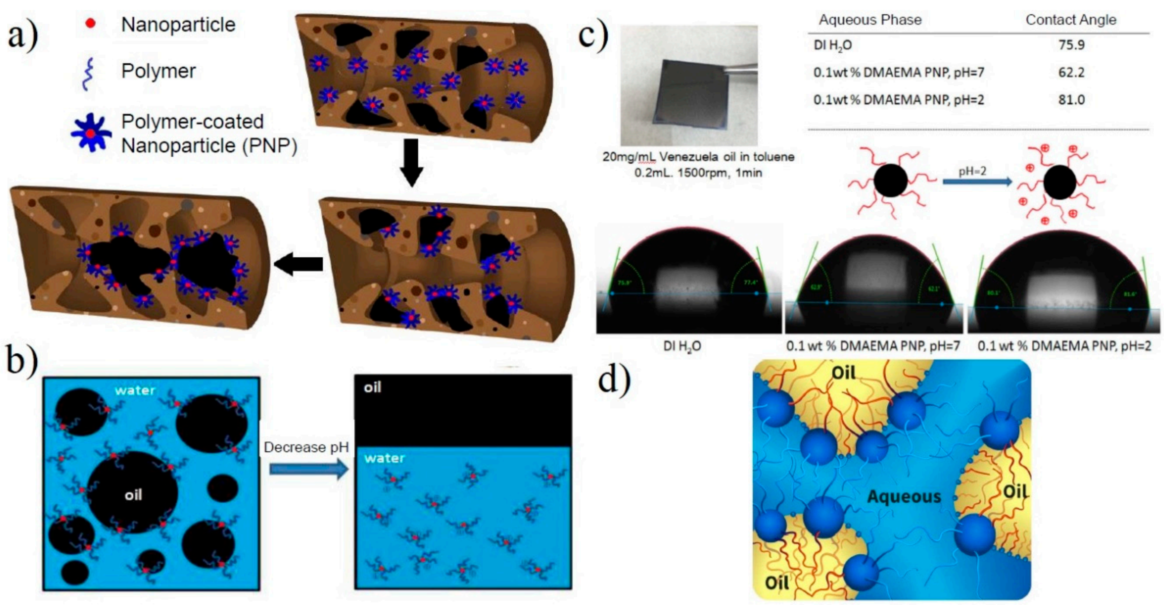

Figure 4. Schematic of (**a**) the use of polymer-coated NP additives to recover heavy oil trapped in porous media, (**b**) the demulsification of oil-in-water emulsions with changes in solution pH, (**c**) wettability alteration by polymer-coated NPs [40] and (**d**) schematic of oil–water microemulsion phases with polymer-coated NP additives [41].

A comprehensive laboratory study on hydrophilic silica dioxide NPs for EOR objectives was carried out at various wettability levels and temperatures of reservoirs (oil-wet, intermediate-wet and water-wet) by Hendraningrat et al. [42]. Relationships between the temperature, initial wettability and additional oil recovery and the nanofluid flooding (nano-EOR) process were investigated. Experimental results have shown that NPs have stable conditions at high temperatures and avoid the aggregation of particles in porous media. Collectively, their results reviewed nanofluid properties for EOR purposes in a wide range of reservoir conditions.

Recently, some researchers have conducted systematic research about the performance of NPs on surfactant flooding. Zargartalebi et al. [43] studied surfactant flooding with nanoparticles. The results indicated that NPs could improve oil recovery and surfactant flooding in equal NP and surfactant concentrations. In addition, hydrophobic NP-modified surfactants were more efficient than hydrophilic NP-modified surfactants.

In another work, a novel polymer nanocomposite was synthesized by nano-SiO_2 and free radical polymerization for use in a polymer flooding system. Oil displacement efficiency, mobility control ability, salt tolerance, temperature tolerance, viscoelasticity and rheological properties were checked under various conditions. The experimental results showed that polymer nanocomposite as a chemical agent has excellent performance for mobility control ability, temperature tolerance and rheological properties for polymer flooding [44,45].

Hendraningrat [46] also showed that polymeric nanospheres have great potential to recover trapped residual oil from porous media. His study showed that NPs could reduce water permeability and mobility ratio, so polymer solutions move to bypassed oil zones (unswept areas) and cause improved oil recovery. In addition, NPs play a key role in changing oil displacement mechanisms and wettability alteration. The results of flooding with NPs indicated that higher concentrations of NPs increase the recovery of oil more than normal polymer flooding (Figure 5a–d) [47,48]. However, high concentrations of NPs have a low effect on the viscosity and IFT, as well having a negative effect on reservoir permeability due to the blocking of porous media (Figure 5c,d).

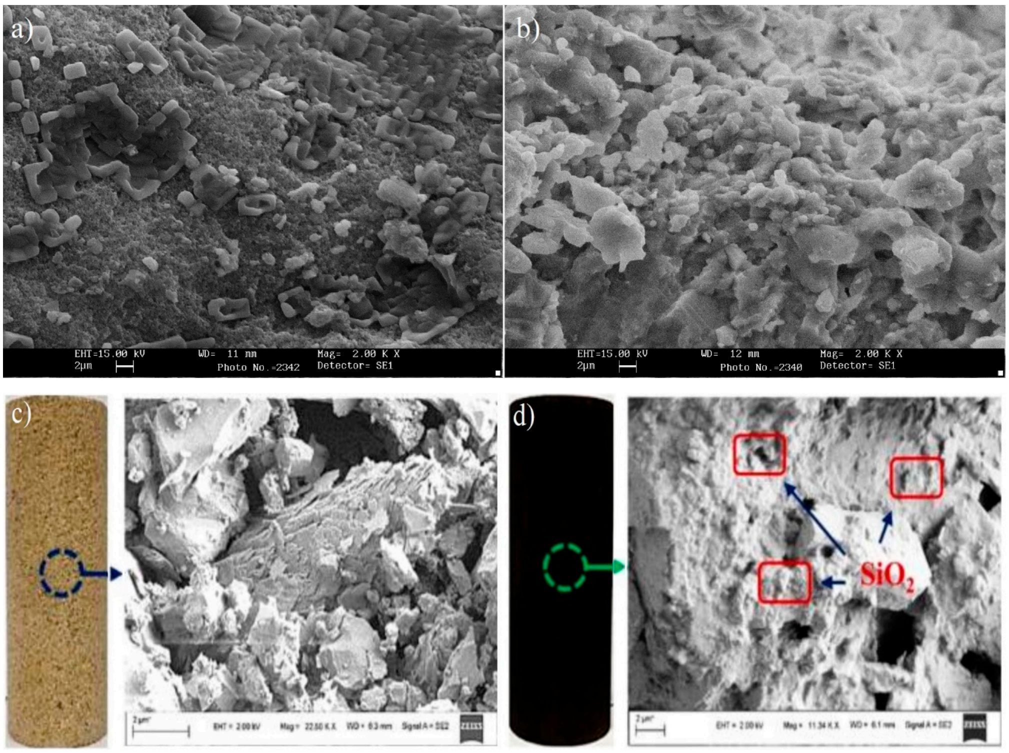

Figure 5. *Cont.*

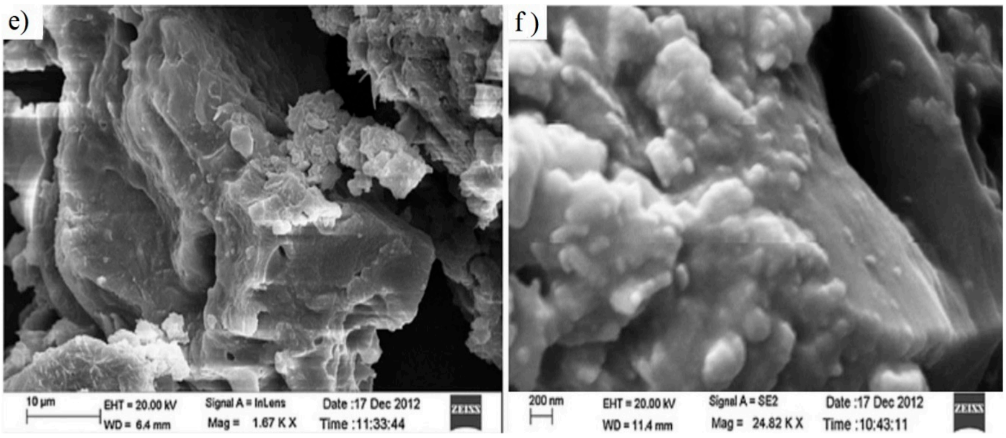

Figure 5. SEM images of sand samples with (**a**) 0.45 wt.% and (**b**) 0.90 wt.% clay NPs after polymer (**c**) Core sample before surfactant–polymer NP flooding, (**d**) deposition of NPs on the core sample after surfactant–polymer NP flooding [48]; (**e**,**f**) SEM images of core plug after flooding with NPs [36].

Some works indicated that hydrophilic silica nanoparticles were distributed through reservoir porous media. According to some research, the concentration of silica NPs should not be more than 0.1 wt.%. This is because the subject can have different effects on EOR. In addition, a high concentration of NPs necessarily result in high oil recovery. However, even a high concentration can decrease residual oil saturation by 2–13% [16]. In a research study, the performance of nano hydrophilic metal oxide was investigated as a polymeric nanofluid for oil recovery by adding polyvinylpyrrolidone. Between NPs, TiO_2 has better performance in polymer flooding and wettability alteration in comparison to silica-based nanofluid [42].

Silica NPs distribute into water-wet porous media but do not move with oil in sandstone channels. Silica NPs with 600 ppm HPAM can, in this system, decrease residual oil saturation by around 20% [24]. Furthermore, propanol and SiO_2 NPs are suitable combinations for oil recovery of around 88% by changing wettability and reducing IFT [49]. Therefore, this subject indicates that NPs can significantly reduce trapped oil if NPs are injected into porous media [49].

The results show that NPs with a 0.1 wt.% concentration cause improved rheological properties in the polymer at a low shear rate (pseudo-plasticity behavior). NPs also cause more enhanced viscosity in nanosuspensions than polymer solutions. In fact, nanosuspensions are a suitable option as they can be applied instead of polymeric solutions to EOR [50].

In order to investigate the performance of NPs and surfactants in sandstone reservoirs, Rahimi et al. [51] studied water-wet NPs (AEROSIL 200) with sodium dodecyl sulfate (SDS) surfactant in a sandstone core. The results showed that injecting NP (1000 ppm) enriched surfactant (2500 ppm) into the core plug causes an 11% increase in ultimate oil recovery. Alteration wettability (from water-wet to oil-wet) by NPs and reduced IFT by surfactant and NPs were the main mechanisms behind this oil recovery. Some experimental research (the effect of nanoparticles on EOR) is shown in Table 1.

Table 1. A summary of types and concentrations of NPs used in EOR.

Type of NP	NP Concentration	Additive	Incremental Oil Recovery	Remarkable Findings	Ref.
SiO_2	1 wt.%	Polyacrylamide	10%	Increased solution viscosity and thermal stability of polymer solution	[52]
SiO_2, $Fe(OH)_3$, TiO_2, Al_2O_3	0.2 wt.%	Xanthan gum	1–9%	Improved solution viscosity	[53]
Al_2O_3	0.05 wt.%	Anionic surfactant	12.5%	Wettability alteration	[54]
SiO_2	0.01–3 wt.%	-	9–19%	Improved oil displacement in porous media	[55]
Clay	0.9 wt.%	Polyacrylamide	5–6%	Improved rheological properties	[56]

Table 1. *Cont.*

Type of NP	NP Concentration	Additive	Incremental Oil Recovery	Remarkable Findings	Ref.
ZrO_2	0.1 g/cc	Cetrimonium bromide surfactant	40%	Wettability alteration	[57]
Pyroxene	1 wt.%	-	10.5%	Reduced IFT and altered contact angle	[58]
Carbon	1 wt.%	-	24.5%	Reduced oil viscosity	[59]
Fumed silica	2 wt.%	Sodium dodecyl sulfate surfactant	13%	Increased solution viscosity	[60]
CeO_2, ZrO_2, Al2O_3, TiO_2, CNT, MgO, $CaCO_3$, SiO_2	5 wt.%	-	8–9%	Increased solution viscosity	[61]
TiO_2	1.9–2.5 wt.%	Sodium dodecyl sulfate surfactant	4%	Improved rheological properties	[62]
SnO_2	2 wt.%	-	22%	Reduced IFT	[63]
ZnO	1.5 wt.%	-	11%	Change in rock wettability, reduced IFT, oil viscosity, mobility ratio and permeability alterations	[64]
Carbon	0.1 wt.%	-	32%	Wettability alteration	[65]
Fluorescent carbon	0.05 wt.%	-	22%	Reduced IFT	[66]
SiO_2	0.3 wt.%	Xanthan gum polymer-surfactant	27%	Increased solution viscosity	[48]
SiO_2, TiO_2, Al_2O_3	0.2–0.4 wt.%	HPAM	7%	Improved rheological properties	[67]
Nickel	0.005–0.02 wt.%	Xanthan gum	6%	Increased solution viscosity	[68]
SiO_2	1 wt.%	Polyethylene glycol	8%	Improved rheological properties	[69]

2.1. Effects of Nanoparticles on IFT

NPs create a thin layer on the surfactant that distributes between oil and injected fluids. This process leads to a significant reduction in the interfacial tension parameter. In fact, in this process, the capillary numbers increase and capillary forces significantly decrease [70]. This means that both surfactant and NPs support each other for injected emulsion stability. In addition, the stability against sedimentation increases in solutions due to NPs that can balance the surface forces and the force of gravity [71–76].

Some researchers claimed that NPs improve surfactant fluids and rheological properties of injection solutions for oil recovery (Figure 6) [77,78]. Figure 6 shows the reduction in IFT between the surfactant solution and crude oil at room temperature along with wettability alteration. In Figure 6, IFT reduction occurs due to NPs' presence between interfacial layers. Surfactant solutions with low NP concentrations cause the absorption process to take place and reduce surface tension. Nonetheless, at high concentrations, nanoparticles nearly cause the depletion of surfactant in the bulk aqueous solution [72,79].

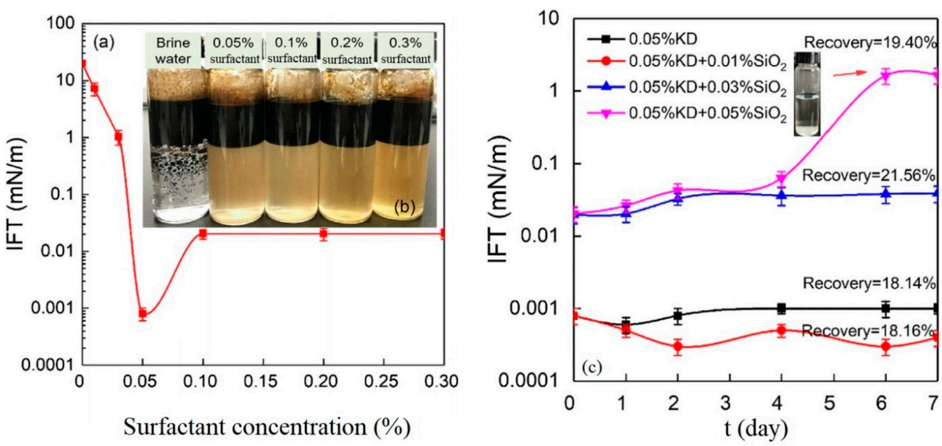

Figure 6. (**a**) IFT between surfactant solutions and oil at 80 °C, (**b**) emulsion stabilization image after 24 h at 80 °C and (**c**) IFT stability of surfactant-based nano-SiO_2 solutions at 80 °C [80].

In order to understand how surfactant and NP additives affect the IFT, Barati et al. [81] used sodium dodecyl sulfate anionic surfactant and NPs (slightly hydrophobic and hydrophilic silica NPs) for modified EOR. The results showed that the value of critical micelle concentration (CMC) plays a key role and can find an optimized amount of nanosuspension surfactant. IFT decreases with the increase in NP-surfactant concentration up to CMC.

In another work, the performance of silica NPs (slightly hydrophobic and hydrophobic) in the surfactant solution was tested [43]. In addition, in order to validate the data, the prepared fluids were investigated in the core flooding test. The results indicated that IFT decreases with increased NP-surfactant concentration, and surfactant adsorption on the surface of the reservoir rock is reduced with an increase in NP concentration. This adsorption reduction was significantly observed in the presence of hydrophobic NPs [43]. In a similar study, the stability of hydrophilic TiO_2 NPs in polyvinylpyrrolidone for the EOR process was investigated. In this work, TiO_2-based nanofluid had excellent performance in wettability alteration and effectively reduced the IFT between the aqueous and oleic phases. Validation of this performance was confirmed with flooding experiments [46]. Some experimental research (the effect of NPs on IFT) is shown in Table 2.

Table 2. Effect of nanoparticles on IFT.

Type of NPs	NPs Concentration	IFT (mN/m)	Reference
SiO_2	0.05 wt.%	17.5	[42]
Al_2O_3	0.5–3 g/L	3.5	[49]
Fe_2O_3	3 g/L	2.7	[49]
CNP	0.1 wt.%	13.4	[66]
SiO_2	1 wt.%	15	[81]
Iron core–carbon shell	100 mg L^{-1}	2.7	[82]
Al_2O_3	0.05 wt.%	23.1	[82]
ZrO_2	0.05 wt.%	6.6	[83]
Hydroxylated nanopyroxene	50 ppm	10	[84]
ZrO_2	10–500 mg/L	2.5	[85]
ZnO/SiO_2	2000 ppm	29.5	[86]
CuO/Fe_3O_4	2000 ppm	4.5	[87]

2.2. *Effects of Nanoparticles on Wettability*

Wettability alteration and its applications are known as the main and most challenging subjects in subsurface engineering. The wettability of reservoir rocks has a considerable effect on the fluid distribution in oil reservoirs and impacts oil recovery during EOR process [88]. Figure 7 is an appropriate description of the effect of wettability on EOR in a micromodel system. The effect of silica NPs on wettability alteration is that this phenomenon causes increased oil recovery. Kuang et al. [89] indicated that a water-wet surface increases the imbibition process into medium and small-sized pores, and lower IFT increases the drainage process and reduces entry pressures (see Figure 7e). Figure 7f–h show brine, simple SiOx nanofluid and complex SiOx nanofluid injection in medium and small-sized pores. The brine has weak performance in displacing oil; however, SiOx nanofluid due to lower IFT produces more oil from medium and small-sized pores.

There are several experimental works in the oil recovery field with hydrophilic silica NPs, most of use silica NPs to achieve wettability alteration. Silica NPs are one of the most used materials for changing wettability. Skauge et al. [24] investigated the oil mobilization properties of nanosized silica particles. They also studied the flow diversion microscopic mechanism by colloidal gel dispersion. In a serial experimental investigation, Hendraningrat used NPs for wettability alteration for use in EOR in reservoirs with different degrees of permeability. In another work, Onyekonwu and Ogolo [24] found that polysilicon NPs (PSNP) have the suitable ability to change rocks' wettability. Their results showed that organically treated hydrophobic and lipophilic PSNP (HLPN) compounds improve oil recovery by more than 50% in water-wet rocks. In similar research, Ju and Fan [90] studied the ability of nanopowders to change wettability for EOR. The results showed that the wettability of reservoir rocks can change from oil-wet to water-wet by the use of lipophobic and hydrophilic polysilicon NPs. In addition, absolute permeability decreases while an improvement in effective permeability occurs. In another study by Hendraningrat [42], IFT and contact angle were analyzed to investigate displacement mechanisms in a chemical EOR process. The results showed that nanofluid with 0.05 wt.% of hydrophilic NPs changes wettability by between 15% and 33%, while NPs had no considerable effect on IFT reduction. Moreover, it was observed that hydrophilic NPs (in an optimized range) with wettability alteration have an effective role in oil displacement in porous media.

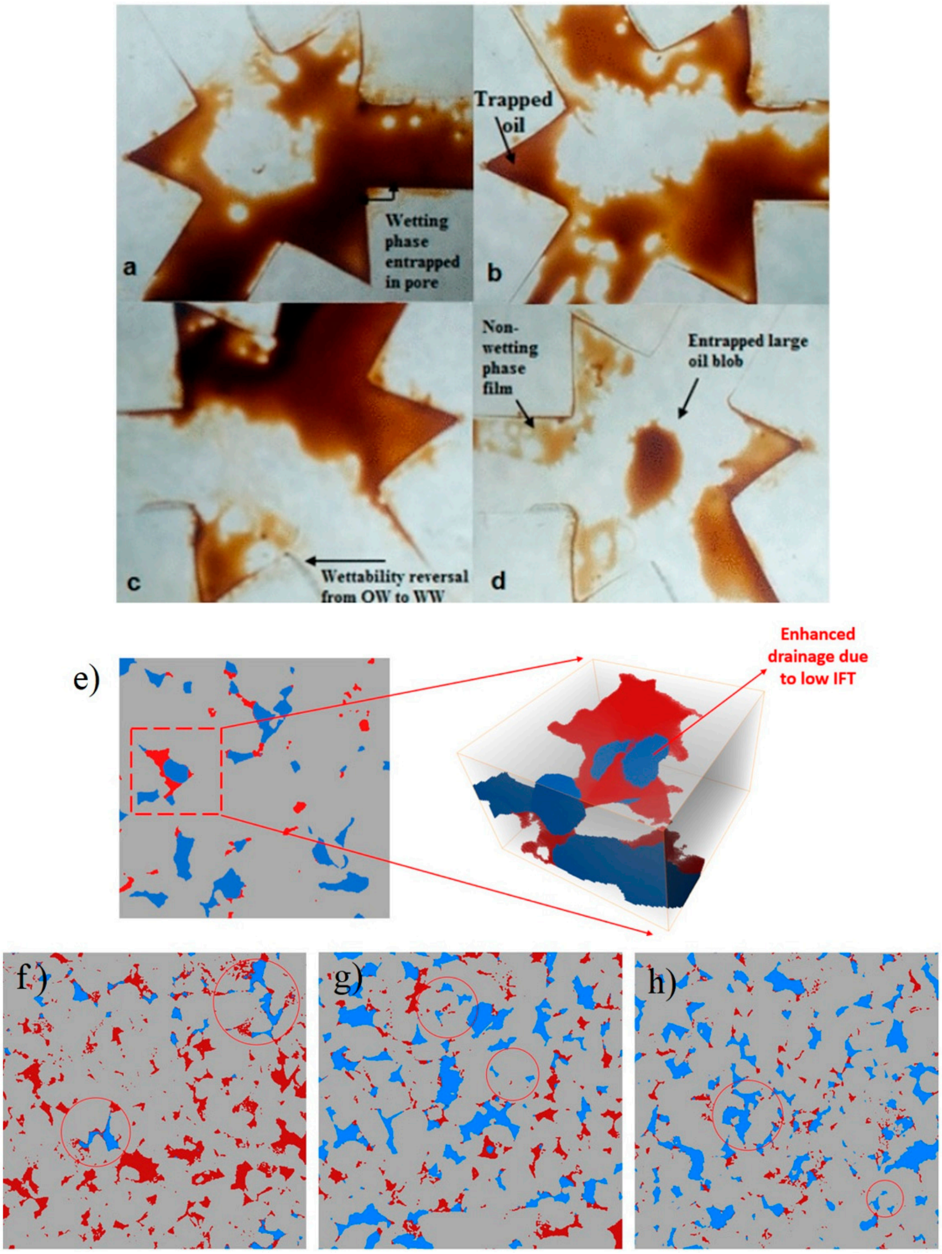

Figure 7. Digital microscope images of solution injection in the micromodel showing the pore-scale configuration and distribution of wetting and non-wetting phases within an initially preferential oil-wet medium for (**a**,**b**) surfactant solution and (**c**,**d**) surfactant and NP solution [60]; (**e**) 3D illustration of enhancement of the drainage process after a complex nanofluid injection and (**f**–**h**) 2D cross-sectional fluid occupancies for nanofluids in oil-wet systems [89].

NPs are also smaller than colloidal particles and have great potential for use in the EOR process. The droplets of emulsion NPs are smaller than colloid, preventing them from becoming trapped, allowing them to easily pass from holes and pores of the reservoir rocks. Commonly, spherical fumed silica particles are mostly used due to the short average diameters (less than 10 nm) and can change the wettability of the surface through silanol groups. At least 10% of silanol groups need to be coated with silica NPs in order to create hydrophobic properties [91–93]. Between NPs, zirconium dioxide has great potential to change the surface wettability during surfactant flooding and create a water-wet surface [94–96]. Silica NPs reduce trapped oil in throats and pores and increase heavy oil recovery from reservoirs [97]. In addition, silica NPs are a suitable choice for changing wettability in polymer flooding, as well as causing increased sweep efficiency and heavy oil recovery. However, the experimental results indicated that other NPs have greater wettability alteration ability and IFT in the case of light oil (Figure 8) [98]. Other research shows that some hybrids such as silica-multi walled carbon nanotube (MWCNT) have excellent ability to change the wettability and improve the rheological properties of injected fluids in reservoirs. The results of Ershadi [99] showed that the wettability of carbonate and sandstone rocks rapidly changes from oil-wet to water-wet by using the MWCNT-silica nanohybrid.

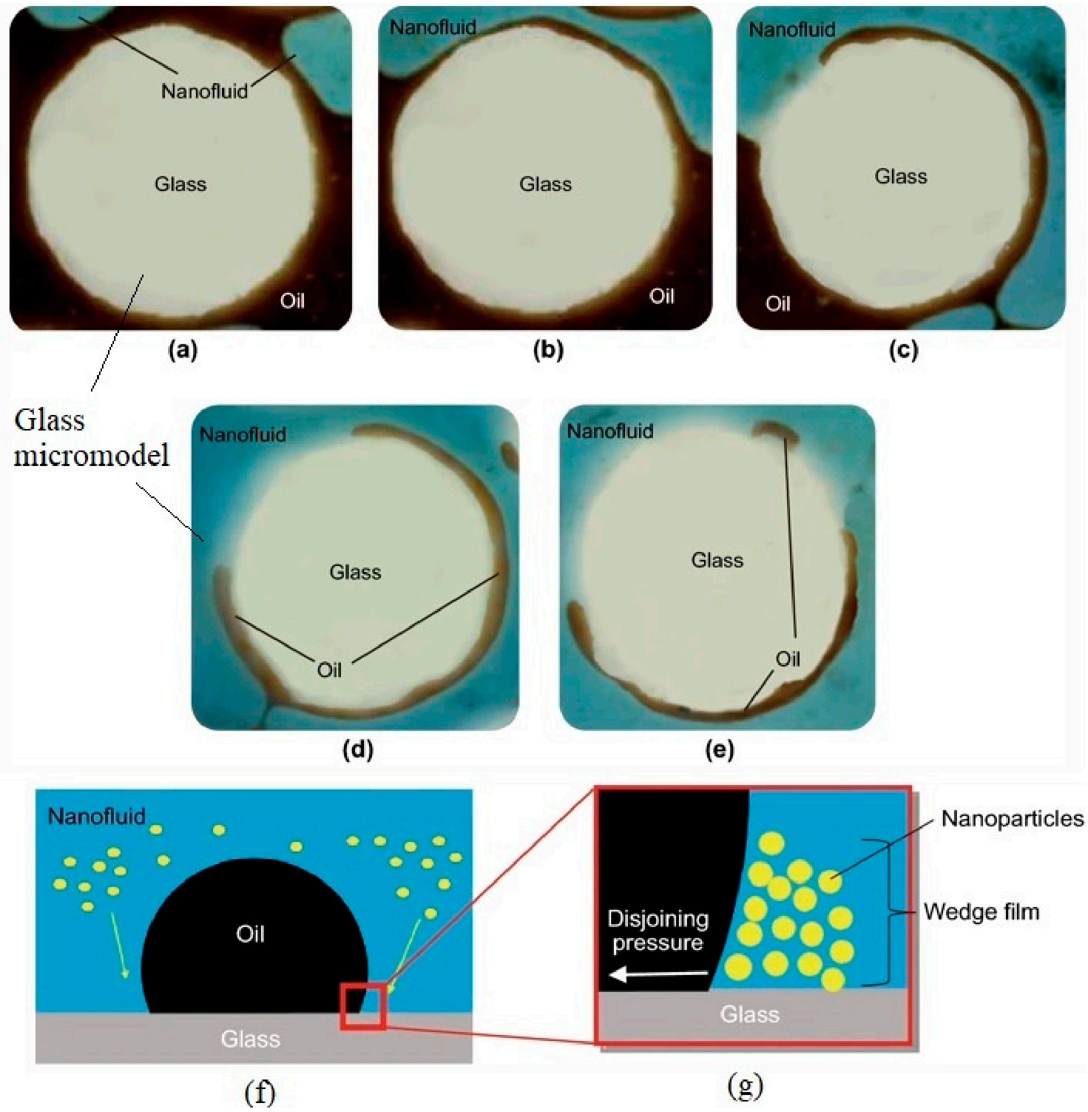

Figure 8. Surface wettability changes. Pore-scale views of the micromodel during nanofluid flooding for a period: (**a**) initial time, (**b**) 1000 s, (**c**) 2000 s, (**d**) 3000 s, (**e**) 5000 s of nanofluid injection; (**f**,**g**) illustration of oil droplet being loosened from the glass surface by adsorption of silica NPs [98].

In other studies, unsteady-state displacement tests of water/water saturated on a sandstone core sample by dispersed nanoparticles–light crude oil systems were performed. The results showed that NP additives have a stronger effect on changing the nonwetting phase relative to permeability curves in comparison to the wetting phase in both drainage and imbibition processes [99–104]. Some experimental research (the effect of NPs on contact angle) is shown in Table 3.

Table 3. Effect of nanoparticles on contact angle.

Type of NP	NP Concentration	Initial Contact Angle (°)	Difference in Contact Angle (°)	Reference
SiO_2	0.05 wt.%	29	28	[42]
Al_2O_3	0.5–3 g/L	134	39	[49]
Fe_2O_3	3 g/L	134	31	[49]
Al_2O_3	1000 ppm	141	32	[54]
Fumed silica	2.2 wt.%	102	92	[60]
MgO	0.005 g/ml	39	42	[61]
CNP	0.1 wt.%	35	85	[66]
Al_2O_3	0.05 wt.%	110	70	[82]
Iron core–carbon shell	100 mg L^{-1}	108	76	[82]
ZrO_2	0.05 wt.%	70	10	[83]
Hydroxylated nanopyroxene	50 ppm	76	52	[84]
ZnO/SiO_2	2000 ppm	132	98	[86]
CuO/Fe_3O_4	2000 ppm	135	61	[87]
γ-Al_2O_3	0.5 wt.%	80	11	[105]
SiO_2	0.05 wt.%	39	28	[106]

3. Challenges and Future Research

Even though the use of NPs can bring significant technical benefits, the drawbacks concerning the use of NPs in EOR cannot be ignored. NP cost is an important issue which should be considered before starting each project. Generally, the synthesis, service and production process of NPs can be very expensive; moreover, large injection volumes of NPs may be required during the EOR operation. Different conditions and unique properties of each oil field and well and the compatibility of NPs with them are other important challenges in this issue. High temperature, chemical alterations and salinity in some formations can be destructive agents for NP structures. Therefore, the use of standard industrial hygiene may significantly improve the safety hazard protection in oil and gas operations with NPs. Finally, since nanotechnology is a relatively new technology in upstream industries, there were limited field tests in this field, so it needs to develop more and more in the pilot stage and oilfield conditions. Therefore, more attention to the numerical and simulation investigation, development and investigation of the effects of different NPs in EOR, as well as eco-friendly methods for the synthesis of nanofluids for use in oil recovery, are now required.

4. Summary

Nanotechnology can be a collection of technically effective options for EOR methods in oil reservoirs. Some mechanisms have been proposed for improving oil recovery with nanoparticles. The effects of NPs and nanofluids on IFT, rheological properties and wettability alteration are the most important mechanisms in this process. The use of NPs as additives in polymer and surfactant solutions can change the rock wettability, reduce the IFT and improve the rheological properties. Finally, though the future boundaries of nanotechnology are completely unclear, nanotechnology will certainly revolutionize and develop the oil and gas industries.

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Nomenclature

Abbreviation	Full Form
NPs	Nanoparticles
IFT	Interfacial tension
EOR	Enhanced oil recovery
LPG	Liquid petroleum gas
PSNP	Polysilicon NPs
HLPN	Hydrophobic and lipophilic polysilicon nanoparticles
Na-Mt	Na-montmorillonite
HPAM	Hydrolyzed polyacrylamide
SEM	Scanning electron microscope
SDS	Sodium dodecyl sulfate
CMC	Critical micelle concentration

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