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Review Article

Application of nanotechnology for enhancing oil recovery – A review

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A B S T R A C T

Nanotechnology has attracted a great attention in enhancing oil recovery (EOR) due to the cost-effective and environmental friendly manner. The size of nanoparticles for EOR usually is in a range of 1–100 nm, which may slightly differ from various international organisations. Nanoparticles exhibit significantly different properties compared to the same fine or bulk molecules because of much higher concentration of atoms at their surface as a result of ultra-small size. In particular, one of the most useful and fascinating properties of these particles is to creating a massive diffusion driving force due to the large surface area, especially at high temperatures. Previous studies have shown that nanoparticles can enhance oil recovery by shifting reservoir wettability towards more water-wet and reducing interfacial tension, yet this area is still open for discussion. It is worth noting that the potential of nanoparticles to reduce the oil viscosity, increase the mobility ratio, and to alter the reservoir permeability has not been investigated to date. Depending on the operational conditions of the EOR process, some nanoparticles perform more effectively than others, thus leading to different levels of enhanced recovery. In this study, we aim to provide a summary on each of the popular and available nanoparticles in the market and list their optimum operational conditions. We classified nanoparticles into the three categories of metal oxide, organic and inorganic particles in this article.

1. Introduction

Extensive research in Nanotechnology commenced in the 1980's and since then it has continued to be a modern wonder of scientific discovery. The nanoscopic Scale is the boundary between Quantum and Bulk effect. Therefore, classical physics and quantum laws can only explain the behavior of these nanoparticles to some degree. However, the reason for this type of behavior and majority of the details of the characteristics of nanoparticles remains a mystery. In nanoscale, particles perform entirely different from their larger scale counterparts. Scientists relate the cause of these bizarre behaviors to the quantum effect or the higher density of atoms at the larger surface area of these particles. These unique properties of nanoparticles have expanded their application in modern life. The oil and gas industry has been closely involved in this change and is still exploring the benefits of this technology. Only recently the high potential of this technology in the upstream oil and gas industry has been discovered. One of the main target areas is to investigate the possibility to improve all conventional EOR methods applications by injecting nanoparticles. Engeset [1] in his work observed that these tiny particles can penetrate into the pore spaces that conventional recovery techniques simply are not able to do so and, therefore, resulted in higher recovery. He also explained that nanoparticles can be tailored to alter reservoir properties such as wettability, improve mobility ratio, or control formation fines migration. Since then a significant amount of research has been devoted to this area. However, there is still a great deal to be explored and learned. This article attempts to summarize the significant results of the research that has been

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completed so far in the area of application of nanoparticles in EOR. What comes next is an attempt to summarize the results of the research conducted to date on the potential applications of nanoparticles in the area of EOR.

2. Nanoparticles and EOR

Currently, known nanoparticles are classified as "fullerenes, graphene, carbon nanotubes, quantum dots, polymeric particles, metallic and metal oxides." [2]. However, not all of these known nanoparticles have been researched for their potential EOR applications. As stated earlier, this article mainly focuses on the concept of nanotechnology application in the oil and gas industry, particularly in EOR. Therefore, in the context of EOR, this paper subdivides the nanoparticles into the three categories of metal oxide, organic and inorganic particles.

2.1. Metal oxides nanoparticles

Clugston and Flemming [3] in their book explained that metallic elements form more than 75% of the periodic table. They include groups 1 to 12 of the table that embraces alkali metal, alkaline metal earth and transition metals plus lanthanides and actinides at the bottom of the table. The main properties of these elements include low ionisation potential, low electronegativity, shiny surface, high melting point and high density. Low ionisation potential and low electronegativity makes them very reactive and unstable elements. They effortlessly lose electrons in a reaction. Therefore, they have a tendency to react with oxygen when they come in contact with it and reach to a stable state. Kuznetsov [4] in his book writes about using the electronegativity to characterise the molecules and their chemical bonds. He defines polar or hydrophilic molecule as the molecule with a large enough difference in electronegativity of the atoms of the molecule. On the other hand, if this difference is small the molecule is non-polar or hydrophobic. Murov [5] explained the updated general guideline of electronegativity differences of the bonded elements that consider a molecule with the electronegativity difference of 0.5–1.7 as a polar covalent or hydrophilic and a molecule with a difference smaller than 0.5 as non-polar or hydrophobic. Based on these definitions metal oxides are always considered as a polar molecule or hydrophilic.

2.1.1. Aluminum oxide (Al_2O_3) NP

Ogolo et al. [6] explained that Al_2O_3 nanofluid is capable of reducing the oil-brine IFT and oil viscosity. However, the oil viscosity reduction is the dominant effect, especially when the particles are dispersed in brine. The results of the conducted core-flood in sandstone with this nanofluid showed that the highest recovery achieved was when brine or distilled water was chosen as a dispersing agent. The researchers recommended this nanofluid as an EOR agent for heavy oil reservoirs. Ogolo et al. [6] also separately investigated the spontaneous imbibition recovery in sandstone cores that showed the highest recovery when Al_2O_3 nanoparticle was dispersed in diesel. Later Hendraningrat and Torsæter [7] researched the emulsion stability of 0.05% wt Al_2O_3 when dispersed in 3% NaCl wt% brine and its effect on oil recovery factor. Their initial measurement showed that Al_2O_3 has the closest specific surface area to titanium (TiO_2) and silica (SiO_2) nanoparticles comparing to other metal oxide nanoparticles. The experiments conducted by these researchers showed that the solution of this nanoparticle in brine was not stable, and it started to precipitate in the first hour. To minimise this problem, they added Polyvinylpyrrolidone (PVP) also known as Povidone with a chemical formula of (C_6H_9NO) to the solution. Results revealed that the PVP successfully stabilized the emulsions. In the second section of their experimental work, the emulsion stabilities of Al_2O_3, TiO_2, and SiO_2 nanoparticles were observed while increasing the temperature up to 80 °C. Interestingly, the results revealed that the aggregation behavior of the emulsions to be almost independent of the temperature. Finally, the core flooding tests conducted by Hendraningrat and Torsæter [7] using the Al_2O_3 nanofluid on Berea Sandstone cores with different wettability at room temperature resulted in the highest recovery for the intermediate wet, the lowest recovery for oil-wet and an in-between recovery for the water-wet rocks. They reported the wettability alteration as the dominant mechanism despite of the fact that it reduced the IFT value as well. Hendraningrat and Torsæter [7] reported Aluminum Oxide, as a good agent for EOR in a sandstone reservoir.

2.1.2. Copper(II) oxide, CuO NP

The reported cases of the usage of this nanoparticle as an EOR agent in literature is very limited. The main research about the potential of this nanoparticle for EOR was conducted by Shah [8]. He used CuO nanoparticles in the laboratory to enhance the recovery of heavy oil with an API gravity of 15 from Berea Sandstone when subjected to gas injection. The results showed that the CuO nanoparticles with the size of 23–40 nm could reduce the heavy oil viscosity as well as increase the density and viscosity of the injected gas (i.e. CO_2). Furthermore, Shah found that adding only 1% of the CuO nanoparticles to the mixture of surfactants (Poly-Di-Methyl-Siloxane (PDMS)) and CO_2 used during EOR, could increase the nanofluid's viscosity significantly (by 140 times). Therefore, since this nanoparticle increased the viscosity of the injection fluids while decreased the viscosity of the oil, Shah concluded that CuO nanoparticles are more efficient when used in the recovery of heavy oils. Through the viscosity measurement, Shah recorded the lowest oil viscosity at 30 mol% CO_2, PDMS and 1% wt CuO nanoparticles at 122 °F. This mixture recovered 71% of the OOIP. Later on, Shah also attempted to increase the nanoparticles' concentration to 3% wt to see if the recovery could be improved further. However, the results showed that the oil viscosity had increased with increasing the concentration from 1% wt to 3% wt.

2.1.3. Iron oxide, (Fe_2O_3/Fe_3O_4) NP

These nanoparticles have unique magnetic properties that make them very useful for applications in magnetic and electrical fields such as sensors, imaging, and data storage. Wu et al. [9] have stated that the main known magnetic molecules from this group are "Fe_3O_4 magnetite, $FeIIFeIII_2O_4$, ferromagnetic, superparamagnetic (Size < 15 nm), a-Fe_2O_3 (hematite, weakly ferromagnetic or antiferromagnetic), c-Fe_2O_3 (maghemite, ferrimagnetic), FeO (wüstite, antiferromagnetic), e-Fe_2O_3 and b-Fe_2O_3". Kothari et al. [10] hypothesised that Iron oxide particles mainly work by reducing the viscosity. After conducting some experiments, Ogolo et al. [6] suggested that iron oxide, when dispersed in brine, performs as a reasonably good EOR agent in sandstone reservoirs. They repeated the same investigation with ethanol as the dispersing agent with no satisfactory results. Furthermore, the studies performed by the same researchers on spontaneous imbibition in sandstone rocks showed that when Diesel was selected as a dispersing agent, Iron Oxide can be a legitimate EOR candidate with instances of 82.5% total oil recovery. It is worth noting that the experimental conducted by Ogolo et al. [6] also revealed that aluminum oxide and nickel oxide were more effective than iron oxide in improving the oil recovery.

2.1.4. Nickel oxide (Ni_2O_3) NP

Nassar [11] indicated that this hydrophilic metal oxide, similar to Al_2O_3, can be a good EOR agent for heavy oil recovery. The production process for heavy oil is very challenging. Deposition of asphaltenes during the production of heavy hydrocarbon such as bitumen is one of these challenges. His studies on the effects of dispersed nanoparticles in heavy oil showed the recovery of up to 85% of the asphaltenes in the original solution. Later Ogolo et al. [6] conducted spontaneous imbibition and core flood experiments on sandstone rock samples at room condition. They observed that in the case of spontaneous imbibition experiments, aluminum, nickel and Iron oxides were good EOR agents especially when diesel was selected as the dispersing agent. In the case of the core flooding experiments, nickel oxide nanoparticles were found to increase the oil recovery when injected into sandstone cores after water flooding. The recovery factor was found to be higher particularly when brine was used as the dispersant for nickel oxide particles. The study claimed that the nickel oxide nanoparticles are capable of increasing the viscosity of the displacing fluid and decreasing the viscosity of the displaced oil.

2.1.5. Magnesium oxide (MgO) NP

Huang el al. [12] investigated the application of nanoparticles to control fines migration in reservoir formation. In their study, they used a particular type of coated crystalline MgO nanoparticles that had been mentioned previously in the literature by its commercial name of nanoparticle 20/40 US mesh. The results obtained by these researchers using this metal oxide are promising. However, MgO nanoparticles effects on EOR requires greater investigation. Later on, other researchers [6] showed that magnesium oxide and zinc oxide used during core-flood tests while dispersed in brine or ethanol can cause permeability impairment in sandstone rocks. On the positive side, Ogolo et al. [6] found that soaking the rock samples in ethanol and magnesium oxide nanoparticles solution could reduce the oil viscosity significantly. This study concluded that overall, MgO nanoparticles are weak recovery agents for EOR in sandstone reservoirs.

2.1.6. Tin oxide (SnO_2) NP

Based on the investigation performed by Najel [13], tin oxide nanoparticles have recently attracted a lot of attention from researchers in various fields. Its unique properties makes it suitable for "n-type semiconductors, transparent conductive electrode for solar cells, gas sensing material for gas sensing devices, transparent conducting electrodes, photochemical and photoconductive devices in liquid crystal display, gas discharge display, lithium-ion batteries, etc.". Najel [13] also listed the currently known process methods for producing,SnO_2 Nanoparticles as "spray pyrolysis, hydrothermal methods, chemical vapor deposition, and thermal evaporation of oxide powders and sol–gel method". Jiang et al. [14] were able to produce tin oxide nanoparticles as a result of heating a mixture of ethylene glycol solutions and $SnCl_2$ at atmospheric pressure. Later, Patil et al. [15] indicated that synthesis of SnO_2 nanoparticles is also possible through a simple hydrothermal route in the attendance of the surfactant hydrazine at 100 °C for 12 h. Generally, SnO_2 has not been used in EOR processes extensively. Studies were done by Ogolo et al. [6] about the potential of SnO_2 as an EOR agent. The results obtained by these researchers showed that SnO_2 performs similar to zirconium oxide and increases the oil recovery in sandstone cores while dispread in distilled water. It is worth noting that the tests mentioned above were conducted at room temperature. Also, the results achieved by Ogolo et al. [6] showed that using ethanol or brine as dispersing agents decreases the recovery factor.

2.1.7. TiO_2 NP

Ehtesabi [16] has indicated that water flooding tests using TiO_2 nanoparticles could recover about 80% of the oil from oil-wet Berea sandstone in an EOR process. He explains that this amount was only 49% in the absence of these nanoparticles. The side measurement performed by Ehtesabi [16] did not reveal considerable change neither in viscosity of the fluid nor the interfacial tension of the fluid system with the use of TiO_2 nanoparticles. Therefore the higher recovery could not be explained with any of these two mechanisms. However, contact angle measurements revealed the ability of TiO_2 nanoparticles to alter the wettability of the sandstone cores from oil wet to water wet. This wettability alteration caused by the deposition of the particles on the rock's internal pore surfaces. SEM (Scanned Electron Microscope) observation confirmed the tendency of TiO_2 nanoparticles to precipitate on the pore surfaces of the rock. Therefore, this study finally found the wettability alteration caused by the deposition of nanoparticles as the main mechanism of additional oil recovery. In another study, Hendraningrat and Torsæter [7] demonstrated that when TiO_2 nanoparticles were dispersed in 3% wt NaCl brine it didn't form a stable emulsion and started precipitating in the first hour and therefore adding a stabilizer was necessary. Povidone as a stabilizer agent with 1 wt% concentration was successful in stabilizing the emulsion. However, these researchers recommended further investigation to identify the optimum stabilizer and its concentration. One of the interesting observations of this research was that increasing the temperature did not influence the aggregation behavior of the particles. As explained by Hendraningrat and Torsæter in their paper, the tendency of particles to aggregate and precipitate resulted a milky solution in water that made measuring the IFT impossible. However, the conducted core floods at room temperature on different wettability Berea sandstones showed that it can recover more amount of oil than SiO_2 in intermediate-wet and less amount oil than Al_2O_3 in oil-wet formations. This study's final conclusion was that, when used as an EOR agent, TiO_2 nanoparticles reduce the oil-brine IFT however wettability alteration is the dominant mechanism.

2.1.8. Zinc oxide (ZnO) NP

Feng [17] in his book explains that ZnO can have polar and non-polar structure. He relates the a-plane ZnO film to non-polar and c-plane to polar. As indicated by Jelinek [18], ZnO nanoparticles have been used considerably for applications such as stabilizers for rubber material, ceramics, food, semiconductors, ointments and photocatalysis. There has been some ongoing research on a particular shape of ZnO called ZnO nanorods (NRs). Feng [17] further explains that NRs shows better physical properties compared to ZnO nanoparticles. Investigations about the applications of this inorganic compound in EOR processes have been very limited. Ogolo at al. [6] explain that, similar to magnesium oxide, when ZnO was implemented as an EOR agent in sandstones, it negatively affected the permeability of the samples used. Evidently, this was more significant when brine or ethanol were used as dispersing agents. The study claimed that the problem initiated by agglomeration of the zinc oxide nanoparticles at the injection point can block the pores. Therefore, due to this problem zinc oxide nanoparticles could lower the overall oil recovery factor.

2.1.9. Zirconium oxide (ZrO_2) NP

As explained by Myloslavskyyl [19] zirconia nanopowders have recently been applied extensively in the industry. He listed these applications as catalysis, ceramics, thermal barrier coating, solid oxide fuel cell components, drug delivery, microelectronics such optical storage and stereo television glasses, high-temperature and corrosion resisting components, temperature and pressure transmitters, transmitting elements, electrode used for magnetic-current generator, heating elements and etc. However, this nanoparticle is not common in the oil and gas industry particularly in EOR process. In 2012, Ogolo et al. injected this metal oxide as an EOR agent at room temperature into a sandstone core sample. The results showed a small increase in oil recovery compared to injection of distilled water alone. On the other hand, when brine or ethanol were used as dispersing agents, it actually reduced the recovery factor to less than that achieved in the absence of nanoparticles.

2.2. Magnetic nanoparticles

2.2.1. Ferro nano-fluids

For the first time, Kothari et al. [10] introduced the idea of using Ferro Fluid, referred to as “smart-nano fluid”, in combination with surfactants in EOR related processes. However, the research was only limited to studying the rheological properties of the Ferro Fluid and did not investigate the use of this fluid in EOR processes directly. In this research, the application of the surfactant was to coat the ferromagnetic nanoparticles to avoid agglomeration. This study claimed that this technique works by reducing the IFT and can be used in both oil-wet and water-wet reservoirs. It is further explained by Kothari et al. [10] that the addition of a surfactant would significantly reduce the interfacial tension and, furthermore, in water-wet reservoirs where isolated oil bubbles are trapped in the center of the pores, Ferro Fluid makes the bubbles to collapse. Furthermore Kothari et al. [10] added that due to the presence of dipole moment, the reservoir fluid molecules align, thus reducing the resistance to the flow that subsequently leads to higher recovery. However, the possibility and practicality of this claim still have not been investigated yet. Recently Huh et al. [20] registered their patent on using hydrophobic magnetic, paramagnetic, or super-paramagnetic nanoparticles incorporated in the fluid as intelligent crude oil tracers. These particles have a formula of XY_2O_4, where X and Y are metal atoms, and X, Y or both are Fe.

2.2.2. Cobalt ferrite NP

In 2012, for the first time [21], investigated the application of cobalt ferrite ($CoFe_2O_4$) nanoparticles in EOR. They synthesised these particles through the sol-gel method. Two separate experiments were conducted. In the first experiment, these particles were applied to magnetic feeders with an antenna that improved the magnetic field in the experiment, and in the second experiment they were applied as nanofluid. In their experiments, cobalt ferrite nanoparticles were added to nanofluid and injected into a sample constructed by packing glass beads in a PVC column. Subsequently, this sample was exposed the EM (electromagnetic) waves for 24 h, at 55 °C. Compared to the case where the nanofluid were injected on its own, this method decreases the residual oil from 31.58% to 8.70%. The study related this increase in recovery to the absorption of electromagnetic waves by cobalt ferrite nanoparticles that decreased the oil viscosity. Sodium Dodecyl Sulphate (SDS) with 1% concentration was used as the stabilizer. This particular percentage is chosen because of its ability to create the highest suspension layer in the emulsion. Despite the above satisfactory results, not much research has been done using these nanoparticles since then, and more investigation is required using core-flood apparatus, in particular, to determine if similar results can be achieved (see Fig. 1).

2.3. Organic nanoparticles

In this article, the word organic refers to the traditional definition that embraces all the compounds that contain carbon in their structure.

2.3.1. Carbon NP

According to America Eelement online glossary [23] carbon is the sixth element in the periodic table and one of the most abundant elements in the universe. This black powder consists of spherical shape nanoparticles with unique properties that generally are synthesised through a hydrothermal process. These particles can be desirably surface modified, with organic molecules or polymers chemically bound to the particles surfaces. A study was conducted by Yu et al. [24] using Berea Sandstone and dolomite cores at room temperature and in the presence of salt ions for the purpose of comparing the breakthrough times of water and various type nanomaterial. The results of this study revealed that the breakthrough time and retention when nanoparticles were used were adversely affected by salt ions. Also, the study concluded that the retention can be improved by controlling nanoparticle surface properties by using different coatings. The retention was higher for dolomite cores because of the attraction between the negatively charged nanoparticles and the positively charged surface of the dolomite core. Therefore, in positively charged formation in carbonate reservoir using surface modifications is highly recommended. For the purpose of determining the suitability of carbon nanoparticles for harsh reservoir conditions Kanj et al. [25] conducted an extensive research using the challenging conditions of a carbonate reservoir called ArabD, with reservoir temperatures greater than 100 °C and high salinity formation water (120,000 ppm). The outcome of this research was the design of a new modified carbon nanoparticle formula commercially known as A-Dots. These particles are carbon based fluorescent nanoparticles and are considered as an example of carbon nanoparticles family group. Based on the results achieved in the above mentioned research, the application of these particles in carbonate samples increased the oil recovery factor to more than 96%. Carbon nanoparticles are only one of the many types of nanomaterial made out of carbon atoms. Among others diamond nanoparticles, fullerene-C60(Buckeyballs), fullerene-C70, fullerene C76, fullerene C78, fullerene C84, graphene, graphite nanopowder have attracted the most attention. Carbon nanotubes are

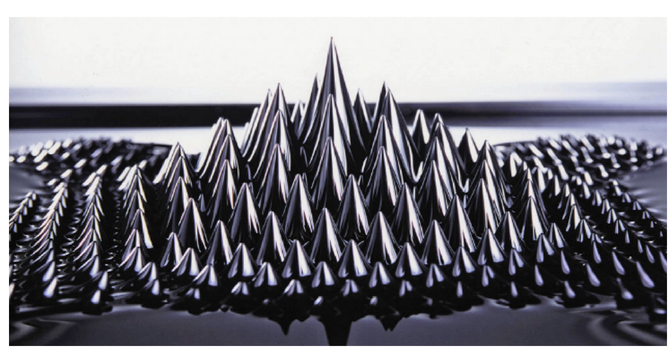

Fig. 1. The magic of ferrofluid in the presence of electromagnets Courtesy [22].

the second member of this family and their potential use for EOR has only recently come to the notice of scientists.

2.3.2. Carbon nanotubes (CNT) NP

Carbon nanotubes are good candidates for building various instruments since they are light, strong and resistant to corrosion. Donaldson et al. in their book [26] classified these nanotubes under Fullerene family. The nanotubes can be single or Multi-walled. Each wall is made from graphene. Each carbon atom in this wall forms three sp^2 hybridised bonds and extra electron from each carbon atom can move through the atom network. Therefore, these tubes exhibit astonishing properties regarding electrical, mechanical and thermal conductivity. These nanotubes are enormously hydrophobic, and thus they have a non-wet water surface. Toluene is a good solvent for carbon nanotubes. There have not been many studies done on using the CNT itself as an EOR agent. However, Chandran [27] carried out a research study on the potential use of Multiwall Carbon Nanotubes (MWNT) fluid as an EOR agent for high-temperature and high-pressure reservoirs. The core-flooding tests were conducted in two different ways, first in the absence and then in the existence of electromagnetic waves. The result of the first test type showed 36% oil recovery after injection of the MWNT nanofluid. The assistance of electromagnetic fields in the second test type almost doubled the recovery. The higher results have been directly related to the oil viscosity reduction associated with the electromagnetic field. Also, the application of these nanotubes has been reported to increase the efficiency of drilling fluids [28] (see Fig. 2).

2.4. Inorganic nanoparticles

In this article, the word inorganic refers to the traditional definition that embraces all the compounds that are lacking carbon in their molecular structure. This group is further subclassified to particles with and without silica.

2.4.1. Silica containing nanoparticles

2.4.1.1. SiO_2 NP. In the book published by the Society for Mining, Metallurgy and Exploration [30] it is explained that silicon dioxide also known as silica is one of the most abundant compounds on earth and is the principal constituent of the sand and sandstone. Therefore, this has made silica to be one of the most commonly used and cost effective nanoparticles. Silica can be obtained naturally from quartz or can be created synthetically. Experimental studies done by Ogolo et al. [6] on the application of SiO_2 in water-wet sandstone reservoirs showed that it could be considered as a suitable EOR agent for this type of rock. In the presence of ethanol as a dispersing agent, it altered the wettability to intermediately -wet in addition to IFT reduction. Wang et al. [31] through their research revealed that the specific surface area of the SiO_2 powders barely changes even when they are heated to various temperatures up to 650 °C. This proves the SiO_2 has good thermal stability. Hendraningrat and Torsæter [7] suggested that Silica nanoparticles form a more stable emulsion in 3%wt NaCl brine compared to metal oxides and does not need a stabilizer. The results achieved by Hendraningrat and Torsæter (2015) showed that this mixture, despite having a higher oil-brine IFT compared to a mixture of brine and stabilizer on its own, resulted in higher oil recovery from Berea sandstone. This proves that the wettability alteration from water wet Berea sandstone to intermediately -wet is the dominant mechanism for oil recovery. By analysing the various literature, it becomes apparent that although under specific circumstances such as the use of water-wet or intermediate wet rock cores. The use SiO_2 nanoparticles during core-floods conducted at room temperature resulted in less recovery, however, generally, it is still considered as a suitable EOR agent in all different wettability conditions from water wet to intermediate and oil wet. In 2011, Nhu et al. studied the synergistic blends of SiO_2 nanoparticles and surfactants for EOR in high-temperature and high brine hardness sandstone reservoirs. They performed experiments combining different types of anionic surfactants with SiO_2 nanoparticles. Some of the blends showed high potential for EOR application because of their resistance to adsorption onto the rock surface, thermostability as well as IFT reduction to ultra-low values. Miranda et al. [32] have discussed some other benefits of using silica nanoparticles for EOR purposes. As described by the authors, one of the advantages is the ease with which the physical chemistry properties of this inorganic compound could be influenced. Also, using silanisation with hydrophilic hydroxyl groups, hydrophobic sulphonic acid, and hydrophilic polyethylene glycol, the surface property of SiO_2 can be switched between hydrophobic and hydrophilic states. Furthermore, Miranda [32] added one main drawback in using this nanoparticle is its tendency to aggregate to larger sizes. Subsequently, this may prevent them from flowing through the pores and pore throats inside a rock (typically measured in microns) and may block the pores. Therefore, the stability of this nanofluid especially in high salinity and high temperatures is always a concern while used as an EOR agent. In Mirandas' et al. studies the emulsion properties of SiO_2 nanoparticles were measured at 300 K and 0.1 MPa pressure and ($CaCl_2$ and NaCl) 1 wt% salinity. Miranda et al. observed the diffusion coefficient increased by increasing salt concentration. The oil/nanoparticles IFT measurements showed the lowest value for hydroxylated nanoparticles and highest value for Functionalized PEG nanoparticles. However functionalized PEG Nanoparticles caused the higher

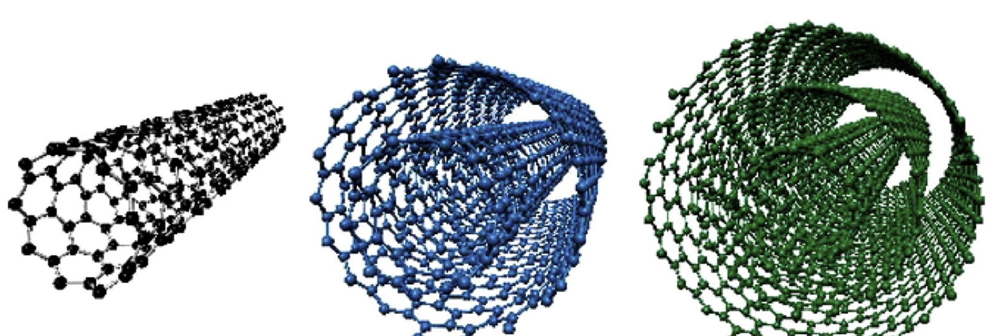

Fig. 2. Ball and stick illustrations of single (left), double (center) and triple-walled carbon nanotubes [29].

nanoparticles mobility. This was because of higher repulsion with the double band of aromatic ring and also higher contact angle between the nanoparticles and the oil/brine interface, which overall led to higher oil recovery (see Fig. 3).

2.4.1.2. Alumina coated silica NP. Hoeck et al. (2011) explained that alumina coating on the SiO_2 nanoparticles entirely alters their properties. The coating creates a positive charge on the surface of this nanoparticle. The Result of their work showed that Alumina coated SiO_2 nanoparticles have a higher surface area compared to the bare SiO_2 nanoparticles however if released into the environment, they show lower toxicity than bare SiO_2 nanoparticles at concentrations ≥46 mg/l, except at pH 6.0. At low concentrations, no clear pH effect was observed for alumina coated SiO_2 nanoparticles. While at higher concentrations phosphate insufficiency could have caused the higher toxicity of those particles at pH 6.0–6.8 compared to higher pH values. Over years, many researchers have used a nanoparticle with the commercial name of LUDOX® CL colloidal silica (Sigma-Aldrich) in their EOR related studies. However, in one study [35] the Propyl Gallate (PG) (Sigma-Aldrich) is used to modify the surface area of the 20 nm nanoparticles to make them partially hydrophobe. The study revealed that the alumina coated silica nanoparticles with modified surface formed a more stable foam and could recover more oil from sandstone cores compared to nanoparticles or surfactant floodings alone on their own (see Fig. 4).

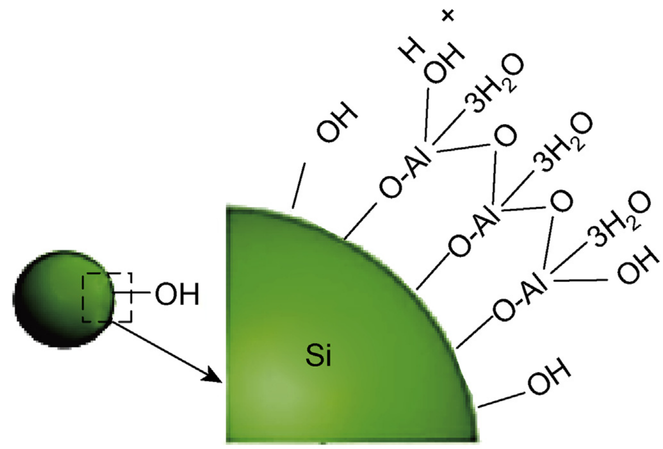

Fig. 4. Sketch of alumina coated nanoparticle [35].

2.4.1.2.1. Hydrophobic silicon oxide (SiO_2) NP. Salyer [36] demonstrated that one common way of synthesizing silica is the addition of silanol (Si-OH) groups on the surface of the Silica nanoparticles which creates hydrophilic particles, originally. Later, if these particles are processed further through a chemical reaction with other reagents, hydrophobic silica particles could be produced. Often the required further process involves adding hydrocarbon group coatings such as alkyl or dimethyldichlorosilane and hexamethyldisilazane chains to the silanol group. Furthermore Salyer [36] classified hydrophobic silica nanoparticles as fumed silica, precipitated silica, and aerosol assisted self-assembly. Studies completed by Ogolo et al. [6] showed that this nanoparticle can be a good EOR agent for sandstone reservoir when ethanol is used as the dispersing

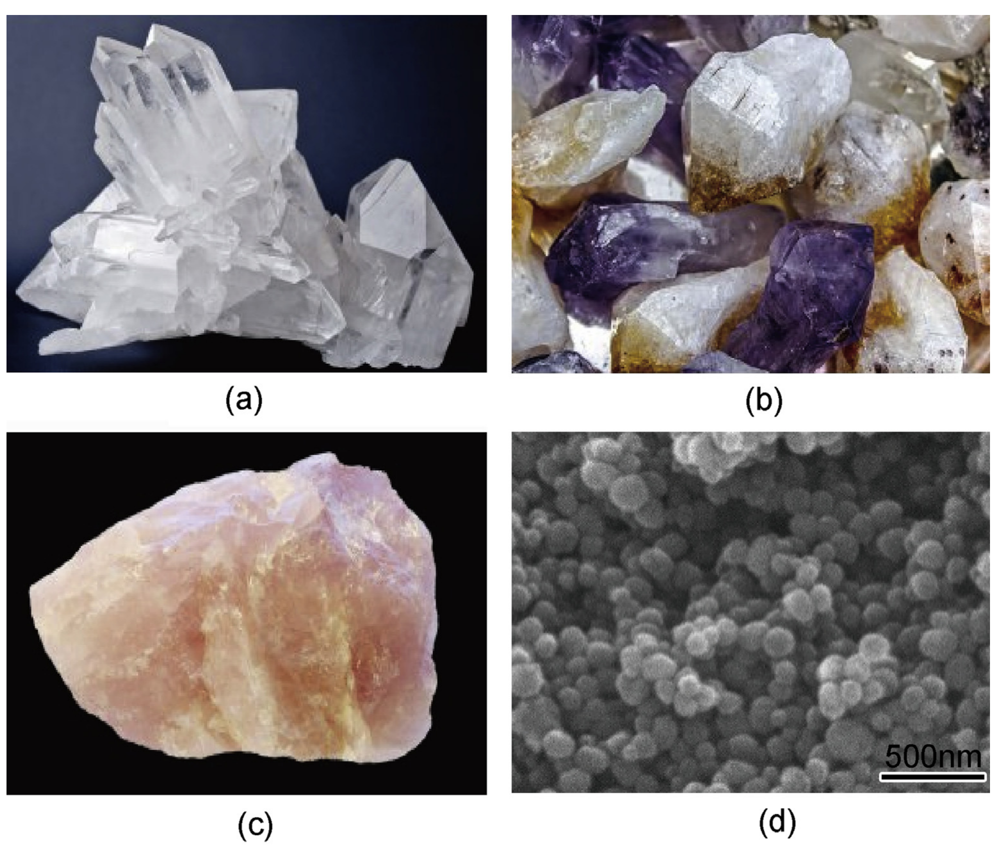

Fig. 3. From left a) Quartz in the most abundant mineral found on Earth, b) Amethyst is a colored type of quartz, c) Rose quartz is often used in jewelry, (a, b and c, courtesy [33]) d) SEM image of mesoporous silica: courtesy [34].

agent. The recovery factor was higher for this nanoparticle comparing to aluminum oxide, magnesium oxide, iron oxide, nickel oxide, zinc oxide, zirconium oxide and tin oxide. Zhang et al. (2010) suggested that these nanoparticles form stable water in oil emulsion and the contact angle at water-oil interface is greater than 90 °C The viscosity of the water in oil emulsion decreased with increasing the salinity from 0 to 10 NaCl wt% and there was an opposite trend for the hydrophilic nanoparticles emulsion. For the unit volume of stable emulsions, the hydrophilic nanoparticles emulsions which are oil in water produced a higher amount of oil comparing to water in oil emulsions made by hydrophobic nanoparticles. For hydrophobic nanoparticles increasing the nanoparticles concentration from 0.1 to 5 wt%, dispersed more water with a smaller droplet in the emulsion.

2.4.1.3. Spherical fumed silica NP. Zhang et al. [37] defined these particles as the most frequently used type of nanoparticles as a stabilizing agent for oil/water emulsions. The wettability of the surface of the particles depends on the degree of coating using silanol groups. High degree of coating (greater than 90%), helps to forms a hydrophilic surface around the particles that is necessary for creating a stabilized oil-in-water emulsions. On the contrary, covering only 10% of the surface with the silanol coating, does not disturb the hydrophobic behavior of the surface and makes the particles suitable for the water-in-oil emulsion. Partially covering the surface creates nanoparticles with intermediate hydrophobicity. To the best of the authors' knowledge, the application of this nanoparticle in EOR studies has been limited to using them as stabilizing agents and its direct application in other EOR related work such as flooding experiments, has not been studied yet. However, the small size of these particles in the range of several to tens of nanometers reduces the risk of blocking the pore in an EOR process.

2.4.1.4. Inorganic silica-core/polymer-shell nano composite. These nanoparticles are SiO_2 nanoparticles in the core that have been covered with a shell of synthetic polyacrylamide polymer. Nguyen et al. [38] showed that these nanoparticles with a composite nature, are suitable for high temperature and high salinity applications in the presence of hard ions commonly found in offshore reservoirs. The particles are efficient in reducing the IFT, increasing the viscosity at critical concentrations. They also have high thermostability and salt tolerance. Based on the results obtained by Nguyen et al. (2012), injecting mixtures of 200 ppm core-shell nanoparticles and 800 ppm blends of two surfactants (anionic and nonionic) in a fractured granite sample at 92 °C and 3.44 wt% salinity could increase the oil recovery by 6.2%.

2.4.1.5. Silicon oxide treated with silane NP. Different types of silane functional group can be found in the market. Jian-Shu (2009) used aminopropyltriethoxysilane, 3-glycidoxypropyltrimethoxysilane and 3- methacryloxypropyltrimethoxysilane to modify SiO_2 particles that resulted in "increasing the final monomer conversion, decreasing the particle size, and narrowing the particle size distribution of the poly (MMA-HEMA)/SiO2 composite emulsion". Lin et al. (2011) also have used other silane coupling agents such as glycidyloxypropyltrimethoxy silane (GPTMS), aminopropyltriethoxy silane (APTES), trimethoxysilylpropyl methacrylate (TMPM), and dichlorodimethyl silane (DCMS) to treat the surface-activated nano-silica. Ogolo et al. (2012) have explained that this nanoparticle can be an excellent EOR agent for sandstone reservoirs, especially when dispersed in ethanol. Experiments with this nanoparticle resulted in the highest recovery compared to other nanoparticles such as aluminum oxide, magnesium oxide, iron oxide, nickel oxide, zinc oxide, zirconium oxide, tin oxide and hydrophobic silicon oxide.

2.4.1.6. Polysilicon nanoparticle (PSNP). SiO_2 nanoparticles are the main component of polysilicon [39]. Roustaei et al. in Ref. [40] have done some research about the characteristics of polysilicon nanoparticles. They classified polysilicon nanoparticles into three types of Neutrally Wet Polysilicon Nanoparticles (NWPN), Hydrophobic and Lipophilic Polysilicon Nanoparticles (HLPN) and Lipophobic and Hydrophilic Polysilicon Nanoparticles (LHPN). Measuring IFT and contact angle along with core floods showed that both HLP and NWP nanofluids improved oil recovery through two primary mechanisms of interfacial tension reduction and wettability alteration to the less water-wet condition from the strongly water-wet condition. The study showed that NWP nanoparticles have a stronger influence on rock wettability while, HLP nanoparticles work mainly through a reduction in oil-water IFT. In regards to EOR application, Onyekonwu and Ogolo [41] investigated three different PSNP's that can alter the rock wettability in various ways. The outcome of their experimental work showed that silane treated NWPN, and HLPN treated with a single layer organic compound, could improve recovery over 50% after primary and secondary recovery on a water-wet rock.

2.4.1.6.1. Hydrophobic and lipophilic polysilicon (HLP) NP. Shahrabadi et al. [42] investigated the effects of injecting this nanoparticle in water-wet sandstone cores at room temperature. This study showed that 4gr/l is the optimum concentration of HLP nanoparticles for EOR purposes. The researchers have indicated that the two mechanisms of reduction of IFT and wettability alteration improved the displacement efficiency at pore-scale resulting in enhancing oil recovery during their experiments. But Shahrabadi et al. (2012) believe that HLP nanoparticles have a higher influence on IFT compared with the wettability of the rock. Also, this study claims that the order of injection of the nanofluid and water affects the performance of nanoparticles since when nanoparticles are injected in the beginning of the experiment results in higher recovery compared to with the flooding process during which water is injected first. Onyekonwu and Ogolo [41] have also experimented with this nanoparticle and recommend these nanoparticles mainly for water wet formations to alter the wettability to intermediate water-wet. They also have indicated that the concentrations below 3gr/l can be considered as optimum. These Researchers showed that Ethanol is a good dispersing agent for this nanoparticle that makes the dispersed nanoparticles act as surfactants as well. Similar to Shahrabadi et al. in (2012), Onyekonwu and Ogolo (2010) have also concluded that in addition to wettability alteration, this nanoparticle can reduce the IFT significantly. Their experimental results have shown that using these nanoparticles in a water-wet sandstone at room temperature and 2000 psi pore pressure could recover up to 80% of the in-situ oil. Ju & Fan [43] based of their numerical studies recommended this nanoparticle for improving water injection specially in low permeability formation. In addition their experimental results showed the effective permeability of water in the sandstone cores treated with this nanoparticle was improved, while a decrease in absolute permeability was observed. However, further studies are required in the future to investigate the effects of temperature and pressure on the performance of these nanoparticles.

2.4.1.6.2. Lipophobic and hydrophilic polysilicon (LHP) NP. Adding an activated additive by γ-ray to the main component of SiO_2, forms a modified ultra-fine powder that is called LHP [39]. In this study, Ju et al. [39] claimed that injection of mixtures of

2.0-3-0% by volume of 10–500 nm LHP nanoparticles in distilled water during core flood experiments could improve the oil recovery. This was believed to be mainly through wettability alteration making the rock samples more water-wet. Ju et al [39] explained further that adsorption of nanoparticles to the pre surfaces of the rock samples caused the hydrophobic pore surfaces to be altered to hydrophilic which accordingly increased the relative permeability of the oil phase (K_{ro}) and decreased the relative permeability of the water phase (K_{rw}), significantly. However, nanoparticles retention has a downside of blocking the pore resulting in reducing porosity and absolute permeability (K) of the porous media. In 2009 another study conducted by Ju and Fan [43] confirmed that adsorption of untreated LHPN can alter the wettability sandstone from oil-wet to water-wet. Other researchers [41] showed that the application of untreated LHPN in water-wet formations is not recommended since it alters the wettability of the water-wet rocks to strongly water-wet and hence could result in a poor recovery factor. The results of the investigations performed to date indicate that this nanoparticle can be a good EOR agent for wet oil formations. Further studies are required in the future to evaluate the effects of temperature and pressure on the performance of these nanoparticles.

2.4.1.6.3. Naturally wet polysilicon (NWP) NP. Shahrabadi et al. [42] explain that similar to HLPN, when using NWP wettability alteration to less water-wetness and IFT reduction are the two dominant recovery mechanisms, especially in water-wet formations. However, NWP nanoparticles have a stronger influence on wettability compared with HLPN. Another study conducted by Onyekonwu and Ogolo [41] claimed that NWP treated with silane are more suitable for water-wet formations. They also recommended that NWP is mainly suitable for reservoirs containing light oil, and its optimum concentration is 3 g/l or less. Later Bagherzadeh (2012) adds that bipolar dispersing agent such as ethanol is suitable for this nanoparticle since it holds both hydrophilic and hydrophobic particles. This study also concludes that NWP can alter the wettability from oil-wet or water-wet to intermediate wet state. Onyekonwu and Ogolo [41] used ethanol as a dispersing agent to make the nanoparticles act as a surfactant as well and therefore, reduce the IFT even further. They concluded that these nanoparticles might not be suitable for intermediate to heavy oil reservoirs. The EOR recovery factor at room temperature and 2000 psi, from water-wet sandstone core samples, using 3% g/L of these nanoparticles dispersed in ethanol ranged from 50% to 70% [44]. Further studies are required in the future to evaluate the effects of temperature and pressure on the performance of these nanoparticles.

2.5. Non-silica nanoparticles

2.5.1. Nano-structured zeolite

Zeolite can be found in either natural form or synthetically made [45]. This type of nanoparticle mainly performs as a catalyst and adsorbent and has been used in refineries and petrochemical plants in cracking process [2]. Studies show that the nanostructured zeolite can extract up to 40% higher amount of gasoline comparing to other catalysts they replaced [46]. Wang et al. [47] were successful in designing a highly selective nanocomposite membranes using Nano crystal-derived hierarchical porous zeolite 4A membrane. The membrane was effective in separating O_2/N_2 molecules in the air. To date, there have been no studies yet on direct application of zeolites in EOR process. However, the potential of ion exchanging nature of zeolite because of its porous structure may need to be investigated. This property of zeolite might be helpful in adsorbing cations such as Na^+, K^+, Ca^{2+}, and Mg^{2+} from the formation water, especially in high salinity conditions. The presence of high concentrations of these cations always is a big challenge in EOR process.

2.5.2. Nano sensors

Kapusta et al. [2] explained that these sensors are in a primary stage. There is still a long way still to achieve a passive nanosensor that does not need external power and possess the ability to store data. However achieving this technology is very beneficial for reservoir exploration. The images from subsurface using nanosensors can provide us with up to 1000 times higher resolution comparing to current methods such as seismic, RF or EM. The combination of the mechanical and acoustic petrophysical data and high-resolution subsurface images and many other accurate information from these nanosensors that are not possible with traditional exploration methods, will enable for a better understanding of the reservoir and using the optimum oil recovery methods. Ragab [46] in regards to the importance of nanosensors wrote that the more we know about the reservoir, the more efficient the process becomes. Implementing nanoparticles sensors is one way that assist us to obtain updated and accurate data from the reservoir by using the advanced measurement techniques.

2.5.3. Nano-Sized Colloidal Dispersion Gels (CDG)

These nanoparticles are commonly used as a sweep improvement agent in EOR process for displacing the oil where the mobility ratio is not desirable [48]. Several studies have also reported the successful experimental application of this nanoparticle in EOR with causing increases in additional oil recoveries up to 10.5% of the OOIP [49,50]. Later Skauge [48] reported that using pre-generated CDG nanoparticles in core flood experiments resulted in up to 40% additional oil recovery. Skauge (2010) attributed this increased recovery to the improvement in the microscopic displacement of the flood. Another core-flood study conducted by Skauge (2010) on water-wet Berea sandstone cores at ambient conditions shows again that pre-generated CDG nanoparticles could result in mobilizing the residual oil left behind from a primary flood. This is something that silica nanoparticles and polymer were not successful in achieving.

2.5.4. Polymer NP

[51] These nanoparticles are either nanospheres or nanocapsules, which are produced via different procedures such as "solvent evaporation, salting-out, dialysis, supercritical fluid technology, micro-emulsion, mini-emulsion, surfactant-free emulsion, and interfacial polymerization." Selecting different methods results in different "particle size, particle size distribution, an area of application, etc." To synthesize a nano-polymer with the optimum properties after choosing the suitable technique, extra attention must be paid to the homogenization procedure, choosing proper surfactants and co-surfactant, and appropriate initiating arrangement. To the knowledge of the author, there has been no work carried out so far regarding investigating the application of nano-size pure polymers in EOR process. The majority of the works so far has focused on polymer composite and polymer coated with organic or inorganic shells.

2.5.5. Polymer coated NP

Johnston et al. [52] used a polymer coating on the surface of the nanoparticle with the usually embedded core of metal oxide to protect the nanoparticle from being adsorbed or other phenomena such as electrostatic interaction. These issues are especially pronounced under the harsh conditions of the reservoir such as high salinity and high temperature. Polyethylene glycol (PEG) is one of the common polymers used for this purpose.

Other researchers such as (ShamsiJazeyi et al., 2014) have showed that polymer-coated nanoparticles (PNP) can be tailored for a particular application through modifying the surface coating. This special feature of PNP has attracted the attention of many researchers, to date. Furthermore (ShamsiJazeyi et al., 2014) added these particles have been used for improving mobility control, altering surface wettability, and foam and emulsion stabilization, reducing IFT and increasing the viscosity of the displacing agents. For further information regarding the application of PNP in EOR, readers are referred to the existing review papers on PNPs. Particularly, the work of ShamsiJazeyi et al. (2014) that is a great summary of the advantages of using these nanoparticles in EOR as well as the challenges such as adsorption and aggregation.

2.5.6. Polyacrylamide Micro-gel nano-spheres

These particles have been used vastly in oilfield EOR techniques. However, their application in EOR was not successful until for the first time Wang et al. [53] were successful in increasing their efficiency to make them suitable for use in EOR processes. They dispersed these nanoparticles in mixtures of two emulsifiers of sorbitan monooleate and polyethylene glycol sorbitan mono-stearate in combination with NaOH. NaOH was the key factor in decreasing the IFT significantly by forming an in-situ surfactants. In the study conducted by Wang et al. (2010), injection of an emulsion containing these nanoparticles was successful in increasing the oil recovery from Zhuangxi sandstone reservoir that contains heavy oil with a viscosity of 238 mPa s at 55 °C. The recovery was increased by over 20% of the OOIP. The ultimate recovery range from 66% to about 78% after injecting nanofluid as a displacing agent. Some of the disadvantages of these particles is the low polymer content and high cost. It is worth noting that the incremental oil recovery factor by nanoparticles usually is below 5% of the OOIP in a saline environment (2 wt% or higher NaCl content) in low concentration (0.01 wt%) [54]. This suggests that there is a pressing need to develop novel nanoparticles to significantly enhance oil recovery with low concentration.

3. Health risks

Archer et al. [55] in their book explain that in more recent years the development in nanotechnology has been so fast that Work, Health, and Safety studies are still trying to catch up. Due to the ultra-small size and unique properties of these particles health risks related to the engineering and use of nanomaterial are not yet clearly understood. Archer et al. further explain that recent studies have found links between nanomaterial and damage to DNA and chromosomes and lung diseases. However, there is still a long way to solid proof of these reported links. This is mainly due to lack of existing measurement equipment in this scale as well as the unpredictable behavior of these particles. Quantum theory and medicine as of today still fails to cover entirely all the grey areas of nanotechnology, when it comes to predicting nanoscale material behavior and their effects on an individual's health. With the minimal available information, long time exposure to nanomaterial could pose a significant risk. Currently in Australia, WHS Act supports the precautionary principle that restricts further research into nanotechnology until there is assurance that it won't endanger people's lives.

4. Conclusion

Nanotechnology has attracted lots of interest recently, there have been various studies that have taken place in regards to the applications of nanoparticle in EOR. This article attempts to summarize and classify the most applied nanoparticles in EOR and to specifically list their dominant mechanisms which leads to enhanced oil recovery.

Viscosity reduction is the dominant mechanism:

- Aluminum Oxide (Al_2O_3)
- Copper (II) oxide, CuO
- Iron oxide, (Fe_2O_3/Fe_3O_4)
- Nickel Oxide (Ni_2O_3)
- ethanol and magnesium oxide
- Polymer Coated Nano Particles

IFT reduction is the dominant mechanism:

- SiO_2 Nanoparticles
- HLP
- Polyacrylamide Micro-gel Nano-spheres
- Polymer Coated Nano Particles
- Ferrofluid

Wettability Alteration is the dominant mechanism:

- Tin Oxide (SnO_2)
- SiO2 Nanoparticles
- Alumina Coated Silica Nanoparticles
- Hydrophobic Silicon oxide (SiO_2)
- Spherical Fumed Silica Nanoparticles
- NWP
- LHPN
- Polymer Coated Nano Particles

Efficient in sweep and displacement efficiency:

- Nano-Sized Colloidal Dispersion Gels (CDG)
- Polymer Nano Particles
- Polymer Coated Nano Particles

Require further investigation in EOR:

- Zinc oxide (ZnO)
- Zirconium Oxide (ZrO_2)
- Ferro Fluids
- Carbon Nanoparticles
- Carbon nanotubes

Concern

- Health & Environment
- Cost
- Aggregation

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