Scope of Implementation of Low Salinity Nanofluid to Improve Oil Recovery in a Part of the Hapjan Oil Field of Upper Assam Basin

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Abstract

The application of nanoparticles in the field of Enhanced Oil Recovery (EOR) has been an emerging technology in recent years. Earlier studies have shown that the nanofluids containing nanoparticles (NPs) of average size (<100 nm) can enhance the oil recovery through wettability alteration of rock, reduction of mobility ratio, reduction of oil-aqueous solution interfacial tension (IFT), reduction of sand production and improvement of sweep efficiency. Low Salinity Waterflooding (LSW) has been utilized as a promising EOR method in the last two decades. Recent studies have found that nanoparticle-assisted LSW embraces both nanoparticles and ions as EOR agents in the injection brine. This study investigates the scope of implementation of the low salinity nanofluid EOR using silica nanoparticles to improve oil recovery in the Tipam Reservoir Sandstone of the Hapjan Oil Field of Upper Assam Basin, India. The analysis of the reservoir brine, reservoir rock and crude oil shows the presence of divalent cations (Ca\(^{2+}\) and Mg\(^{2+}\)), clay minerals (Illite and Smectite) and polar compounds (Asphaltene and Resin) respectively.

The presence of the divalent cations, clay minerals and polar compounds in the Crude Oil/Brine/Rock system of a sandstone reservoir is the prerequisite for implementing low salinity nanofluid EOR. The study shows that the low salinity nanofluids can shift the wettability of the rock to a more water-wet state. It is also observed that the silica nanoparticles can increase the nanofluid viscosity and reduce the oil-nanofluid IFT, which can improve the recovery of oil. The experimental results show that the study area has great potential for the low salinity silica nanofluid EOR to improve oil recovery.

Keywords: EOR, IFT, LSW, nanofluid, wettability.

Introduction

Today global energy demand is growing, while the existing reserves in conventional reservoirs are depleting and the frequency of new hydrocarbon exploration has also been decreasing in the last decades. Additionally, a large amount of hydrocarbon remains unrecovered in the Brown Oil Fields after the primary, secondary and traditional enhanced oil recovery (EOR) methods. It is found that traditional EOR methods constitute around two-thirds of the oil produced from the Original Oil in Place (OOIP)\(^{41}\). As a result, the significance of innovative technology and research is highly understood to maintain the energy demand-supply balance.

In recent years, the use of nanoparticles (NPs) for improving oil recovery is considered as a new emerging EOR technology\(^{48}\). Over the last decade, this technology has been developed greatly and unlocked its potential applications in the field of EOR. The ultra-small size, very high surface-to-volume ratio, low cost and environmental friendly nature of the nanoparticles make them a better EOR agent as compared to the traditional EOR methods\(^{43}\).

It is observed that reduction of formation permeability and loss of chemicals in the pore wall occur during the chemical EOR methods, which results in reducing oil recovery and increasing the cost of injection\(^2\). But, the very small size (<100 nm) nanoparticles like iron oxide (Fe\(_3\)O\(_4\)/Fe\(_2\)O\(_3\)), silicon dioxide (SiO\(_2\)), copper oxide (CuO), aluminium oxide (Al\(_2\)O\(_3\)), magnesium oxide (MgO), zinc oxide (ZnO), nickel oxide (Ni\(_2\)O\(_3\)) etc. can easily flow through the porous media along with the injection fluids as the size of the pore channels and throats are larger than the nanoparticles\(^{46}\). As a result, there is no severe trapping of nanoparticles in the pore throats and reduction of formation permeability. Additionally, they have the ability to improve the sweep efficiency by penetrating some smaller pore spaces of the rock, which is unable to do by the traditional injection fluids.

It is observed that nanoparticles have a very large surface area which increases rapidly with decreasing the diameter of particles\(^{38}\). The large surface-to-volume ratio of the nanoparticles exhibits enhanced diffusion at high pressure and temperature, which improves the recovery of oil\(^{14}\). Also, the cost of the nanoparticles is usually lower than the chemicals used in the traditional EOR methods\(^{43}\). It is observed that some nanoparticles are environmentally friendly in nature. For example, silica nanoparticles contain about 99.8% silicone dioxide, which is the main component of sandstone. Therefore, the most commonly used nanoparticles in EOR are silica nanoparticles due to their environment-friendly nature, low cost and availability\(^{48}\).

It is found that the addition of certain nanoparticles to the injection liquid can significantly improve the oil recovery through one or more different mechanisms. Those
mechanisms are wettability alteration of rock, disjoining pressure, improvement of the sweep efficiency, prevention of asphaltene precipitation, improvement of the mobility ratio, reduction of interfacial tension (IFT), stabilization of foams and emulsions under harsh environments and reduction of sand production\textsuperscript{31,47,48}. The schematic illustrations of some of the oil recovery mechanisms by nanofluids are shown in figure 1.

Alteration of rock wettability from the initial oil-wet or mixed-wet state to a more water-wet state is the most efficient way for the EOR applications. Earlier researchers have shown that nanofluid can change the rock wettability to a more water-wet state\textsuperscript{29}. Nanoparticles present in the nanofluid are adsorbed on the rock surfaces forming a layer of nanoparticles (Figure 1d) that separate the oil droplets from the rock surfaces resulting in higher oil recovery\textsuperscript{3}.

It is also observed that cation exchange (divalent and monovalent) occurs between the nanoparticles and rock surfaces which can result in wettability alteration to a higher water-wet state\textsuperscript{39}.

Earlier studies have shown that the nanofluids can develop a structural disjoining force (film) between the rock and oil surfaces, creating a wedge film structure (Figure 1c) which can increase the desorption of oil from the rock surfaces\textsuperscript{16}. Studies also show that smaller nanoparticles can give higher repulsive forces that can improve the recovery of oil through wettability modification\textsuperscript{20,25}. Silicon dioxide and titanium dioxide (TiO\textsubscript{2}) are the most widely used nanoparticles in EOR which can shift the wettability state to more water wetness by the disjoining pressure mechanism\textsuperscript{13,18,25}.

Due to the constant differential pressure and smaller diameter of pore throats, the velocity of nanofluid increases at the pore throat in a sandstone reservoir. This may cause the nanoparticles to move slower than the water molecules which result in the accumulation of nanoparticles and eventually block the pore channel (Figure 1). This blockage may divert the flow of nanofluid towards the previously unswept area of the reservoir, forcing more oil to produce. Earlier laboratory core flooding experiments have shown that silica nanofluid can improve the oil recovery in a sandstone reservoir by delaying water breakthrough and hence improving sweep efficiency\textsuperscript{38}.

Asphaltene is a polar organic compound of crude oil which remains stable at equilibrium conditions. However, during production or solvent injection into the oil, precipitation of asphaltene occurs from the oil solution and remains suspended in the oil. When the precipitation increases, flocculation of the asphaltene particles occurs which is then deposited on the reservoir rock surfaces\textsuperscript{17}. The absorption of nanoparticles onto the asphaltene can reduce the asphaltene precipitation in the porous media (Figure 1b). The high affinity of the asphaltene to the nanoparticle surface leads to the self-association of the high molecular weight compounds by neutralizing the polar forces which prevent its precipitation and deposition on the rock surfaces\textsuperscript{35}. This reduction of asphaltene precipitation improves the permeability and water-wetness of the rock which in turn reduces the residual oil saturation\textsuperscript{17}.

![Figure 1: Schematic illustration of different oil recovery mechanisms by nanofluids. (a) IFT reduction (b) Preventing Asphaltene Precipitation and Breakdown (c) Disjoining Pressure (d) Wettability Alteration (e) Pore Plugging\textsuperscript{17}](https://doi.org/10.25303/282rjce100109)
One of the most important parameters that govern the performance of a chemical EOR method is the mobility ratio. It is the ratio of the mobility of displacing and displaced phase fluid, which is a dimensionless number. A favorable mobility ratio (<1) is desired for more oil recovery where the displacing phase fluid cannot travel faster than the displaced phase fluid. The mobility ratio can be lowered by viscosity reduction of the displaced phase fluid or viscosity enhancement of the displacing phase fluid. It is observed that nanoparticles can increase the nanofluid viscosity and reduce the oil viscosity which results in a low mobility ratio and hence higher oil recovery. Recent studies have shown that silicon dioxide, aluminium oxide, copper oxide, iron oxide and nickel oxide nanoparticles can increase the NP-water nanofluid viscosity and reduce the oil viscosity.

One of the common strategies to increase oil recovery efficiency is to reduce the oil-brine interfacial tension (IFT). By decreasing the IFT, capillary number can be increased, which in turn improves the microscopic displacement efficiency. Earlier studies have shown that the reduction of IFT is one of the major mechanisms for incremental oil recovery from nanoparticle EOR. This IFT reduction occurs due to the adsorption of nanoparticles at the oil-brine interface that modifies the natural interactions. It is found that silicon dioxide, titanium dioxide, aluminium oxide and nickel oxide (NiO) nanoparticles can reduce the crude oil-brine IFT when their concentrations increase from 0.01 wt.% to 0.05 wt.%. Among all the nanoparticles invested, silicon dioxide is found to be more effective in lowering the oil-brine IFT, whereas nickel oxide generates the highest value of IFT.

Nanoparticles have been used to stabilize foams due to their high thermal stability. Earlier studies have shown that silica and iron oxide nanoparticles have the ability to enhance foam stability in harsh environments for a longer period of time which can reduce the formation damage and improve the plugging performance. Nanoparticles can also stabilize emulsion under a harsh environment which can improve the oil recovery through the mobilization of the emulsified oil blank.

As mentioned above, nanoparticles can alter the rock wettability to a more water-wet state by forming a nano texture on the rock surface. By adsorbing on the rock surface, they also prevent the contact of the rock with the surrounding aqueous solution which reduces sand production. This reduction of sand production greatly reduces the damage to downhole and surface production facilities and also the risk of catastrophic failure.

On the other hand, low salinity waterflooding (LSW) is also a newly developed low-cost and environmentally friendly EOR technique where oil recovery efficiency can be improved by changing the chemistry of the injection brine. The major mechanisms suggested in LSW that lead to more oil recovery include fine migration, pH increase, multicomponent ion exchange (MIE) and expansion of electrical double layer (EDL). The pre-requisite for low salinity waterflooding EOR in a sandstone reservoir is the presence of divalent cations, clay minerals and polar organic compounds in the crude oil-brine/rock system. Earlier studies have found that the presence of plagioclase feldspar in the reservoir rock plays an important role in obtaining low salinity effects. Also, when the colloidal conditions are favorable, mica tends to detach from the rock surface along with the clay minerals and migrate along with the flowing fluid. These migrated fines can get trapped in some pore throats which can lead to the enhancement of sweep efficiency by diverting the injected fluid into un-swept pores.

It is observed that the stability of the nanoparticles in nanofluid is enhanced under a low-salinity environment. Recent studies have shown that silica nanoparticle is more effective in preventing sand production when injected with low-salinity water. Also, silica nanoparticles can reduce the residual oil saturation by modifying the rock wettability and mobility ratio when used along with the LSW. Thus, nanoparticle-assisted low salinity waterflooding embraces both nanoparticles and ions as EOR agents in the injection brine.

In this work, the scope of implementation of the silica nanoparticles in conjunction with LSW has been studied for improving oil recovery in Tipam reservoir sandstone of the Hapjan oil field of Upper Assam Basin, India. Hapjan oil field is a part of the Greater Hapjan Oil Field in the Upper Assam Basin, which is located in the North of Langkasi area of Jorajian Oil Field and Southeast of Makum-North Hapjan Oil Field of India.

**Material and Methods**

For this work, nine numbers of reservoir rock samples were collected from the study area from a depth range of 2315 to 2329 meters. Also, a formation brine sample and 10 liters of crude oil were collected for this study. Silicon dioxide (SiO$_2$) nanoparticles with 99.5% assay were purchased from the Sisco Research Laboratories Pvt. Ltd., Maharashtra, India. Also, sodium chloride (NaCl), calcium chloride (CaCl$_2$·2H$_2$O) and magnesium chloride (MgCl$_2$·6H$_2$O) were purchased from Molytech, Mumbai for preparing the brine solution. The required distilled water was prepared in the Laboratory of the Department of Chemistry, Dibrugarh University Institute of Engineering and Technology, Dibrugarh University, Assam, India.

For this study, eight numbers of low-salinity brine and one formation brine were prepared in the laboratory by mixing NaCl, CaCl$_2$·2H$_2$O and MgCl$_2$·6H$_2$O with distilled water in the required proportions. As mentioned above, the stability of the nanoparticles in nanofluid increases in the low salinity environment. It is also found that a reduction of injection brine salinity by 43% from the formation brine salinity can give the highest oil recovery in the sandstone reservoir of the sandstone reservoir of the
Upper Assam Basin\textsuperscript{27}. Therefore, for this work, the salinity of the low-salinity brines was kept at 570 ppm which is 43\% lower than the formation brine salinity of the Hapjan oil field (1000 ppm). The salinity of the prepared brines was confirmed by Boyle’s method using potassium chromate (K\textsubscript{2}CrO\textsubscript{4}) and silver nitrate (AgNO\textsubscript{3}).

According to Lager et al\textsuperscript{21}, a lower concentration of divalent cations in low-salinity brine than the formation brine is more effective in obtaining low salinity effects. Also, divalent cations (Ca\textsuperscript{2+} and Mg\textsuperscript{2+}) can cause more instability to the silica nanoparticles in the solution as compared to the monovalent cations as mentioned above. Therefore, during the preparation of the low salinity brines, the concentrations of the Ca\textsuperscript{2+} and Mg\textsuperscript{2+} were kept at 32 ppm and 16 ppm respectively which are 43\% lower than their concentration in the formation brine (Table 1).

Table 1

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<tr>
<th>S.N.</th>
<th>Characteristics</th>
<th>Unit</th>
<th>Value</th>
<th>Test Method/Instrument Used</th>
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<td>8.3</td>
<td>Metrohm pH Meter</td>
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<td>2</td>
<td>Calcium</td>
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<td>Complexometric Titration (Using EDTA)</td>
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<tr>
<td>3</td>
<td>Magnesium</td>
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<td>28.8</td>
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<td>4</td>
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<td>Titration with Hydrochloric Acid</td>
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<td>6</td>
<td>Salinity (as NaCl)</td>
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<td>7</td>
<td>Total Dissolved Solid</td>
<td>mg/l</td>
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<td>Hach Water Checker</td>
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</table>

Figure 2: Photomicrographs of the rock samples of the study area showing (A) Quartz, Plagioclase Feldspar, Ferruginous Cement, Polycrystalline Quartz; (B) Quartz, Plagioclase Feldspar, Ferruginous Cement, Polycrystalline Quartz; (C) Quartz, Plagioclase Feldspar, Polycrystalline Quartz, Biotite and (D) Quartz, Ferruginous Cement, Polycrystalline Quartz, Muscovite
The physical properties of the nanoparticles are reported in table 3. A total of seven numbers of SiO₂ nanoparticle solutions were prepared with the low-salinity brines for this study. The nanoparticle concentrations in the nanofluid range from 0.01% (w/w) to 0.5% (w/w). The nanoparticles were mixed with the brine solutions using Ultrasonic Processor (Sonicator, Model EI-750UP).

**Thin Section Analysis:** For this study, a suitable size rock piece was cut from the collected reservoir rock sample with a diamond saw and ground optically flat. The sample was then mounted on a glass slide and ground smooth using progressively finer abrasive grit until the sample is only around 30 micrometers thick. Finally, the section was polished using nylon cloth and diamond paste to obtain a suitable polish surface for microscopic study. The thin section was then studied under the polarized light microscope. In this work, the study shows the presence of Quartz, Plagioclase Feldspar, Ferruginous Cement, Polycrystalline Quartz, Biotite and Muscovite (Figure 2).

**Scanning Electron Microscopic (SEM) Analysis:** The Scanning electron microscopic analysis of the collected rock samples was done to study the presence of clay minerals playing an important role in enhancing oil recovery during nanoparticle-assisted low salinity waterflooding. For this work, the rock samples were analyzed under the Scanning electron microscope JEOL JSM-IT300. The rock samples were coated with gold using sputter coating and placed in the sample chamber. The SEM analysis was carried out at an accelerating voltage of more than 20KV which shows the presence of Smectite, Illite, Feldspar and Mica in the rock matrix (Figure 3).

**X-Ray Diffraction Analysis:** For this study, the grain size of the rock samples was reduced to less than 2 microns using Agate Mortar. X-Ray diffraction analysis was then done with the X-Ray Diffractometer (Rigaku Ultima IV) in the 2θ range of 5° to 60°. The X-Ray diffractogram of one of the reservoir rock samples is shown in figure 4.

**Formation Brine Analysis:** For this work, an analysis of the formation brine of the Tipam Reservoir Sandstone of the Hapjan Oil Field was done to mainly determine the salinity and ionic composition. Earlier studies have shown that salinity and pH of brine govern the stability of the Crude Oil/Brine/Rock system which affects the recovery of oil\textsuperscript{9,44}. Also, the presence of calcium and magnesium ion in the brine plays a vital role in oil recovery during low-salinity nanofluid flooding. The result of the brine analysis is given in table 1.

![Figure 3: SEM photomicrographs of the rock samples of the study area showing (A) Smectite, Illite, Feldspar; (B) Smectite, Illite, Mica; (C) Smectite, Illite and (D) Smectite, Illite, Feldspar.](https://doi.org/10.25303/282rjce100109)
Figure 4: X-Ray Diffractogram of the reservoir rock sample showing Smectite, Illite, Mica-Montmorillonite, Plagioclase Feldspar, Pyrite, Siderite, Hematite and Quartz

Table 2

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<th>S.N.</th>
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<td>API Gravity at 60°F</td>
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<td>20.3</td>
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<tr>
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<td>Pour Point</td>
<td>°C</td>
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<td>pH</td>
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<td>Salinity</td>
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<td>Morh’s Method</td>
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<tr>
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<td>Acid Number</td>
<td></td>
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<td>Designation 139/98 (Under D974)</td>
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**Crude Oil Analysis:** It is observed that the divalent cations (asphaltene and resin) and high acid number of crude oil have a pronounced effect on the oil recovery during low salinity EOR\textsuperscript{12,21}. Therefore, the analysis of the collected crude oil sample was done to determine the different parameters. The results of the crude oil analysis are given in table 2.

**Determination of the Nanofluid Viscosity:** Nanoparticles can increase the mobility ratio by increasing the nanofluid viscosity, as mentioned earlier. For this study, the viscosity of the silica nanofluids was determined using M3600 Viscometer at room temperature. A total of three numbers of nanofluid viscosity measurements were taken for each nanoparticle concentration and their mean value was determined. The variation of the nanofluid viscosity with the silica NP concentration is shown in table 4. The viscosity of the 1000 ppm formation brine was found to be 1.04 cp.

**Determination of the Crude Oil-Nanofluid Interfacial Tension:** As mentioned above, the reduction of oil-nanofluid IFT is one of the major mechanisms that improves the recovery of oil during nanofluid flooding.
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from LSW\textsuperscript{32}. Also, as mentioned above, Mica (Biotite, Muscovite) enhances the oil recovery during LSW through sweep efficiency improvement.

Formation Brine analysis shows that the salinity (as NaCl) is 1000 mg/l, whereas the total dissolved solid is 1241 mg/l (Table 1). The formation brine contains 56 mg/l calcium and 28.8 mg/l magnesium ion. These divalent cations act as bridging agents between the negatively charged oil and clay surface\textsuperscript{23}. When the bulk fluid has a high salinity level, the thickness of the electrical double layer becomes small, due to which crude oil and clays form an organo-metallic complex making the rock surface more oil-wet in nature\textsuperscript{36}. However, during low-salinity brine injection, expansion of the electrical double layer occurs, which results in the wettability alteration of the rock to a more water-wet state\textsuperscript{24}. The presence of divalent cations in the formation brine is also important for the multicomponent ionic exchange mechanism to work during LSW\textsuperscript{21}.

It is found that the crude oil of the study area contains 0.40 % (w/w) asphaltene and 10.14% (w/w) resin (Table 2). As mentioned above, resin and asphaltene (polar compounds) make the rock surfaces more oil-wet in nature by adsorbing on the clay surfaces as organo-metallic complex and/or organic components. But the multicomponent ion exchange mechanism that occurs during LSW can replace the organo-metallic complex and/or organic components with un-complexed cations on the clay surface, which leads to the shifting of rock wettability in a direction of more water-wet state\textsuperscript{5}. The acid number of crude oil is 0.86. According to earlier studies, a high acid number (AN>0.2) of crude oil is important which can generate surfactants in situ to alter the rock wettability to more water-wet during LSW\textsuperscript{11}. Some other researchers also have found that a high acid number (AN>0.2) of crude oil containing resin and asphaltene plays a significant role in wettability alteration of the rock in the direction of more water wetness during LSW\textsuperscript{19,23}.

The study of the variation of low salinity nanofluid viscosity with the concentration of SiO\textsubscript{2} nanoparticles shows that the nanofluid viscosity increases with increasing the nanoparticle concentration at room temperature. With the increase in SiO\textsubscript{2} nanoparticle concentration from 0.01 % (w/w) to 0.50 % (w/w), the viscosity of the nanofluid increases from 1.17 cP to 1.94 cP (Table 4). The increase of nanofluid viscosity by increasing the nanoparticle concentration has also been observed in the earlier studies as mentioned above. This viscosity increase results in a decrease in the mobility ratio which leads to more oil recovery.

Studies have shown that the increase in nanofluid viscosity at higher concentrations is due to the increased molecular interaction between the base fluid and nanoparticles\textsuperscript{6}. The viscosity of the 1000 ppm brine found was found to be 1.04 cP which indicates that the high salinity formation brine viscosity is lower than the low salinity nanofluid viscosity.

The analysis of the effects of SiO\textsubscript{2} nanoparticle concentrations on the crude oil-nanofluid IFT shows that an increase in the concentration of SiO\textsubscript{2} nanoparticles can decrease the IFT. It is observed that crude oil-nanofluid IFT is reduced up to 14.32 mN/m from 24.56 mN/m when the nanoparticle concentration is increased from 0.01 % (w/w) to 0.50 % (w/w) (Table 4). However, the IFT between the crude oil and formation brine is found to be the highest (33.14 mN/m). As mentioned earlier, the reduction of IFT is one of the major mechanisms for incremental oil recovery from nanoparticle EOR (Figure 1a) which occurs due to the adsorption of nanoparticles at the oil-nanofluid interface.

Moreover, the pH increase of the fluid may occur during the injection of low salinity nanofluid, which reacts with the acidic compounds of crude oil. As a result, in situ generation of surfactants occurs which can further reduce the IFT\textsuperscript{9}.

Wettability alteration of the rock is one of the major mechanisms of nanofluid EOR. The present study shows that the contact angle of the rock decreases from 48.28° to 36.21° when the SiO\textsubscript{2} nanoparticle concentration increases from 0.01 % (w/w) to 0.50 % (w/w) (Table 4).

However, the wettability of the reservoir rock in the presence of the formation brine is more oil-wet in nature (Contact Angle 51.24°). This indicates that the wettability state of the Tipam Reservoir Sandstone of the Hapjan oil field is shifted to higher water wetness in the presence of a higher concentration of silica nanoparticles.

Earlier studies have shown that the nanoparticles present in the nanofluid are adsorbed on the rock surfaces forming a layer of nanoparticles (Figure 1d) that separate the oil droplets from the rock surfaces (more water-wet), resulting in higher oil recovery. In addition to this, pH increase, multicomponent ion exchange and expansion of electrical double layer may be the mechanisms which have contributed to the change of the rock wettability to a more water-wet state and in turn more oil recovery.

**Conclusion**

The study shows that the presence of divalent cations in the formation brine, clay minerals and plagioclase feldspar in the reservoir rock and asphaltene and resin in the crude oil have made the study area a suitable candidate for implementing Low Salinity Nanofluid EOR. With the increase in SiO\textsubscript{2} nanoparticle concentration from 0.01 % (w/w) to 0.50 % (w/w) in the low salinity brine, the crude oil-nanofluid IFT and contact angle of the reservoir rock decrease whereas the nanofluid viscosity increases. These low IFT, high water-wet state of rock and high nanofluid viscosity can improve the recovery of oil.

Therefore, the application of SiO\textsubscript{2} nanoparticle assisted low salinity waterflooding in the Tipam reservoir sandstone of the Hapjan oil field of Upper Assam Basin, India has great potential in improving oil recovery.
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