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An experimental investigation of the enhanced oil recovery and improved performance of drilling fluids using titanium dioxide and fumed silica nanoparticles

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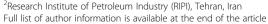
Abstract

Nanotechnology has contributed to the technological advances in various industrial biomaterials and renewable energy production over the last decade. Recently, a renewed interest arises in the application of nanotechnology for the upstream petroleum industry such as exploration, drilling and production. In particular, the adding of nanoparticles to injection fluids may drastically benefit enhanced oil recovery, such as changing the properties of the fluid, wettability alternation of rocks, advanced drag reduction, strengthening sand consolidation, reducing the interfacial tension and increasing the mobility of the capillary-trapped oil. The feasibility of these methods depends on many factors such as flow mechanisms in porous media and porous medium properties at microscopic and macroscopic scales. Previous studies have indicated that the oil recovery from porous media may be substantially increased by the injection of miscible fluids. This all sounds great and waterflooding has been used successfully for decades; however, it is important to carefully design and appropriately operate the waterflood. Using nanoparticles in all samples has resulted in recovery increase. Among these applications of the study is nanoenhanced oil recovery which can be applied in many water-wet reservoirs dominated by inhibition mechanism to extract more fluid through really small caliber pores. In these experiments, two nanoparticles dissolved in water are injected into simulated environment, and also, the effect of these nanoparticles in water-base drilling typical fluid have been investigated. Using nanoparticles in all samples has resulted in recovery increase. Finally, considering the experiments, it is demonstrated that flows with nanoparticles and, in particular, titanium dioxide (TiO₂) nanoparticles have the highest amount of recovery factors and thus using nanoparticles in waterflooding projects and even some in polymer flooding ones. Also, results of the other tests, regarding each typical drilling costs of each foot and importance of time in the operation, it is possible to replace technically and economically the ordinary addition (here, the widely used sodium hydroxide) with fumed silica nanoparticles in drilling fluid to prevent cement contamination of the drilling fluid. The advantages of nano-TiO₂ are possessing suitable thermal transition qualities in the drilling fluid.

Keywords: Nanoenhanced oil recovery (NEOR); Drilling fluid; Porous media; Inhibition mechanism

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Background

In the last few years, enhanced oil recovery (EOR) processes have regained interest from the research and development phases to the oil field EOR implementation. This renewed interest has been furthered by the currently high oil price, the increasing worldwide oil demand, the maturation of oil fields worldwide, and few new well discoveries (Aladasani and Bai 2010 in [1]). Hydrocarbon recovery occurs through two main processes: primary recovery and supplementary recovery. Primary recovery refers to the volume of hydrocarbon produced by the natural energy prevailing in the reservoir and/or artificial lift through a single wellbore, while supplementary or secondary recovery refers to the volume of hydrocarbon produced as a result of the addition of energy into the reservoir, such as fluid injection, to complement or increase the original energy within the reservoir (Dake 1978; Lyons and Plisga 2005 in [1]).

Customized nanoparticles have the ability to enhance oil recovery, improve exploration, and be useful in the formation scale control. Nanoparticles can be tailored to alter reservoir properties such as wettability, improve mobility ratio, or control formations migration. Nanofluids have successfully been developed in laboratories, and the upcoming challenge is to develop techniques for the cost of industrial-scale production of nanofluids. In nearly all cases, the thermal conductivity of conventional heat transfer of fluids is improved by the addition of small amounts of nanoparticles [2]. In addition, development of pressure and temperature-sensitive nanosensors will enable *in-situ* measurements within the reservoir [3]. Nanosensors can provide improved temperature and pressure ratings in deep wells and hostile environments [4].

Many novel enhanced oil recovery methods have been applied to increase oil recovery in oil fields [5,6]. The feasibility of these methods depends on many factors such as flow mechanisms in porous media and porous medium properties at microscopic and macroscopic scales have been reported the role of nanoparticles in EOR operations (Skauge et al. 2010 in [7]). As well, some researchers presented extensive literature reviews regarding the effect of wettability on microscopic displacements of fluid flow at pore scale (Zhao et al. 2010; Jamaloei and Kharrat, 2010 in [8-10]).

Suleimanov et al. (1995 in [11]) carried out experiments which showed how dispersed nanoparticels in an aqueous phase could modify the interfacial properties of a liquid/liquid system if their surface were modified by the presence of an ionic surfactant. The application of nanosuspension in their study permitted significant increase in the efficiency of oil displacement oil-water rate. In homogeneous porous media, oil recovery before water breakthrough was increased by 51% and 17% for surfactant aqueous solution with nanoparticle

addition to water and surfactant aqueous solution, respectively [11].

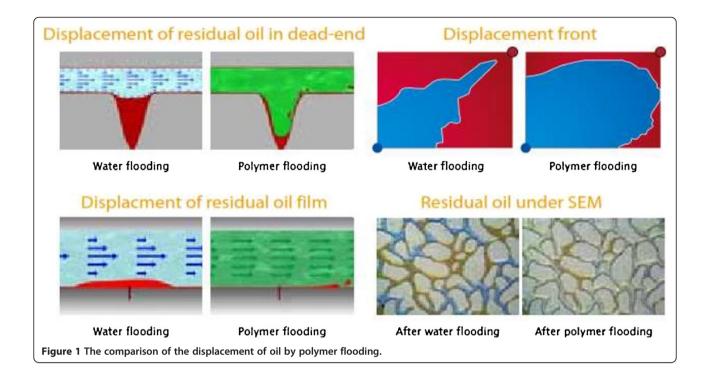
Designed nanoparticles, especially nanocrystalline materials in combination with advanced drilling fluids, will probably improve the rate of penetration and decrease wear on drilling equipment significantly [12]. Nanobased mud additive is expected to improve the thermal conductivity of nanofluids, which consequently will provide more efficient cooling of drill bits, and longer operational cycles [13]. In addition, an additive in casing to increase compressive and exural strength, as well as light-weight, rugged structural materials was studied in the project thesis, The use of nanotechnology in the petroleum industry [4].

Polymer flooding is a tertiary recovery method by adding high-molecular-weight polyacrylamide into injected water to increase the viscosity of fluid, improve volumetric sweep efficiency, and further increase oil recovery factor. When oil is displaced by water, the oil/water mobility ratio is so high that the injected water fingers through the reservoirs. By injecting polymer solution into reservoirs, the oil/water mobility ratio can be much reduced, and the displacement front advances evenly to sweep a larger volume. The viscoelasticity of polymer solution can help the displacement of oil remaining in micropores that cannot be otherwise displaced by water flooding.

Previous studies have indicated that the oil recovery from porous media may be substantially increased by the injection of miscible fluids. This all sounds great and water flooding has been used successfully for decades; however, it is important to carefully design and appropriately operate waterflood. There are many factors to consider when designing a successful water flood including

- reservoir permeability (both absolute and relative)
- beginning and ending fluid saturations (oil, water and gas)
- reservoir heterogeneity
- oil gravity and viscosity
- water source and compatibility
- formation clay content
- depth and lifting costs

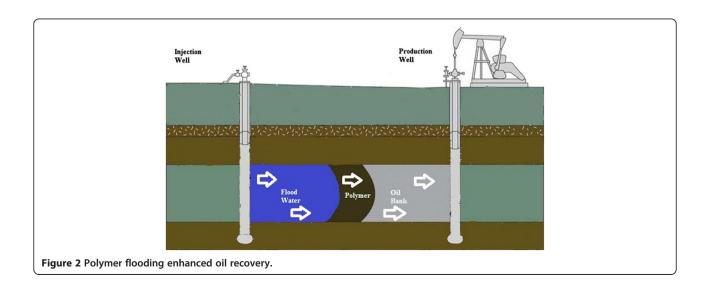
But if done right, a well run water flood will significantly nanoenhance oil recovery and produce attractive returns for many years. Waterfloods can also be improved by polymer flooding. Addition of polymer makes the water more viscous so that oil is produced faster. Obviously, this is not a good idea in a low-permeability reservoir or one with a high clay content that can adsorb the polymer. However, polymer-augmented waterfloods can be profitable. In this experiment, two nanoparticles titanium dioxide and fumed silica dissolved in water are



injected into the simulated environment. Other additive matters that are used in making the compound are introduced in the next parts.

First, using a simulated reservoir, we prepare the container, from whose entrance we inject 100 psi air pressure and through the end there is an opening for flow exit. Here, also a metal net and a filter paper with the same cross sections are used. So, it is possible to simulate an environment like porous media.

Furthermore, it is tried in this paper to study the effect of two nanoparticles in water-base drilling-typical fluid. Specifically, the effect of this matter on the yield point (YP) and consistency index (k) and flow behavior index (n) was examined to improve cement-contaminated cement during drilling operation. Decreasing the flow behavior index by adding nanofumed silica in base fluid is among the results of the test. Cementing operation was of drilling steps employed several times to strengthen casing pipes. So, cement contaminations are avoidable in drilling. Silica (silicate mud) is a drilling fluid that matter used for specific purposes including being economic, lower biocontamination, better thermal



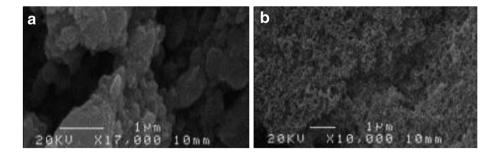


Figure 3 SEM micrographs of the pure oxides (a) TiO₂ and (b) fumed SiO₂.

and pressure stability, lubricating drilling pipes and shale prevention. Regarding each typical drilling costs of each foot and importance of time in the operation, it is possible to replace technically and economically the ordinary addition (here, the widely used sodium hydroxide) with fumed silica nanoparticles in drilling fluid to improve cement-contaminated drilling fluid. One of the nano-TiO2 matter advantages is possessing suitable thermal transition qualities in drilling fluid. In drilling mud composition, there are a series of solid particles making electrochemical bounds with other liquid-phase matters. The resistance the matters show against cut is called yield point the unit; on the other hand, the point expresses viscosity and electrostatic forces between particles. In laminar flow, lines are parallel; mud moves in separate parallel layers with different velocities and passes through ring spaces or pipes. With the increase, flow velocities get closer in different layers. Displacement of mud is more difficult with slow flow because drilling particles tend to layers with low velocity and move in the layers and cause low velocity in the layers, their displacement gets harder. One way to avoid the problem is to increase the yield point.

The designating flow behavior index (n) indicates non-Newton behavior of a fluid, namely, Newton fluids assign n index of unity to themselves; the lower it is, the higher the mud capability in cleaning the well, namely, the wider the longitudinal profile of moving fluid and the lower the solids fall. Numerical reduction of n results in bending lapse of the moving fluid, and as a result, viscosity in the place of drill and drilling speed will increase. Consistency index indicates concentration

Table 1 Composition of the fluids

No. of fluid	Ingredient base fluid (wt.%)	Nanofume silica content (wt.%)	Nano-TiO ₂ (wt.%)	
1	Initial fluid (IF)	0	0	
2	Initial fluid (IF)	0	1.4285	
3	Initial fluid (IF)	1.4285	0	
4	IF + Carboxymethyl cellulose (0.2857)	0	0	
5	IF + Carboxymethyl cellulose (0.2857)	0	1.4285	
6	IF + Carboxymethyl cellulose (0.2857)	1.4285	0	
7	IF + Polyhachioides (0.2857)	0	0	
8	IF + Polyhachioides (0.2857)	0	1.4285	
9	IF + Polyhachioides (0.2857)	1.4285	0	
11	IF + Sulfate solution (4)	0	0	
12	IF + Sulfate solution (4)	0	1.4285	
13	IF + Sulfate solution (4)	1.4285	0	
14	IF + Cement (1.1428)	0	0	
15	IF + Cement (1.1428)	0	1.4285	
16	IF + Cement (1.1428)	1.4285	0	
17	IF + Cement (1.1428%) + Sodium hydroxide (0.2857)	0	0	
18	IF + Cement (1.1428%) + Sodium hydroxide (0.2857)	0	1.4285	
19	IF + Cement (1.1428%) + Sodium hydroxide (0.2857)	1.4285	0	

Table 2 Physical properties of fluids

No. of fluid	Viscosity (cp)	Conductivity (mV)	Mud cake diameter (mm)	Final gel strength	Initial gel strength	Temperature (°C)	Density of fluid (ppg)
1	9	-259	1.9	28	12	22	8.7
2	18	-213	2.71	31	25	22.5	8.7
3	14	-183	2.36	38	13	24	8.8
4	19	-219	1.6	38	18	23	8.6
5	24	-219	1.69	36	20	23	8.7
6	20.5	-161	1.39	52	12	23	8.7
7	11.5	-235	1.98	28	15	31	8.6
8	20	-212	2.56	45	29	22	8.7
9	16.5	-161	3.8	91	34	23	8.8
11	5.5	-177	0.64	6	3	20	8.7
12	5.5	-172	0.6	4	3	20	8.75
13	6	-165	1.31	9	3	20	8.88
14	43	-308	6.28	37	33	23	8.7
15	69	-323	12.48	59	42	23	8.8
16	39.5	-309	8.2	65	33	23	8.6
17	20	-315	9.74	18	12	25	8.7
18	61	-330	7.48	47	36	25	8.6
19	46.5	-337	9.12	73	51	26	8.6

or viscosity and pumping capability of drilling fluid; the higher the viscosity, the higher the k. In case the flow behavior index is low, cleaning the well gets harder and it is possible to increase viscosity gradually to enhance consistency in the index size. Consistency index (k) and flow behavior index change with flow chemical composition and temperature. However, considering relatively constant temperature of the flows under study, the only alternative is changing their chemical composition.

Results and discussion

The comparison of the displacement of oil by polymer flooding is shown in Figure 1, while enhanced oil recovery is shown in Figure 2.

In general, it is observed that for fluids in whose nanostructures or nanoparticles are used (Figure 3), YP is significantly higher than basic fluids. Regarding Tables 1 and 2, it can be seen that fluids YP having ${\rm TiO_2}$ nanoparticles is higher than the other fluids, when sodium hydroxide is used in fluid. YP difference between nanofluids

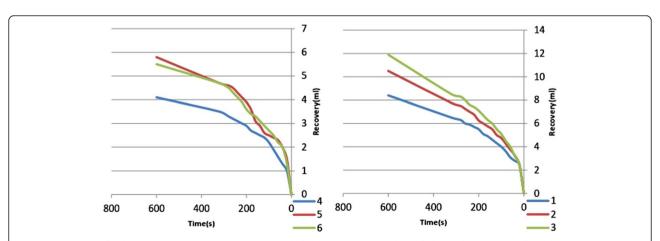


Figure 4 Recovery fluid vs. injection time. Regarding relatively fixed temperature and pH of all samples, we generally analyze them. Samples 1, 2 and 3 have basic fluid and samples 4, 5 and 6 have carboxymethyl cellulose.

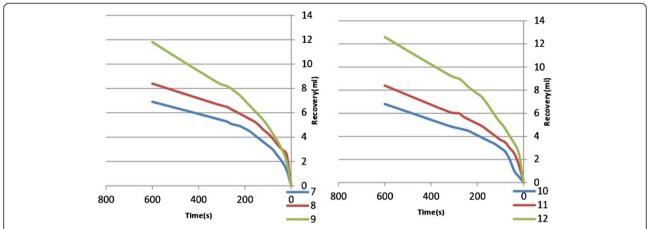


Figure 5 Recovery fluid vs. injection time. Regarding relatively fixed temperature and pH of all samples, we generally analyze that samples 7, 8 and 9 have polyrhachioides and samples 10, 11 and 12 also have 10% sulfate solution.

and basic fluids is considerably higher than the time the matter is not applied in the fluid structure. Also, sodium hydroxide results in higher YP fluid with fumed silica nanoparticles compared to ${\rm TiO_2}$ nanoparticles with fluids. Among fluids, mud has a basic matter and ${\rm TiO_2}$ nanoparticle has a maximum YP. Except fluid having fumed silica nanoparticle their bending tension grows with a constant rate, bending tension of fluids with sodium hydroxide

increase lower than the time the matter present in the compositions. Meanwhile, basic fluid with nano- ${\rm TiO_2}$ without sodium hydroxide has maximum slope of bending tension against bending rate and minimum amount is attributed to the basic fluid with sodium hydroxide relatively determined with a constant slope. Regarding fluids without sodium hydroxide, basic fluid with fumed silica nanoparticles and higher YP compared to the basic fluid,

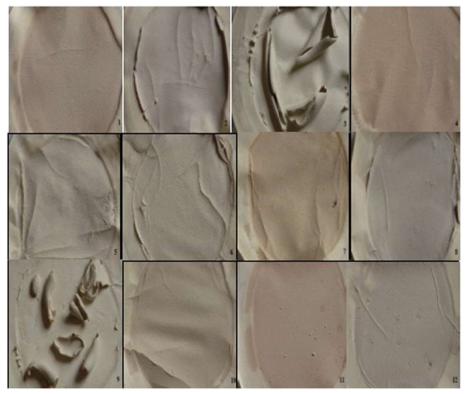
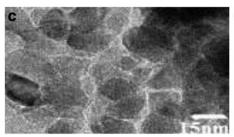


Figure 6 Effect of TiO₂ and fumed silica nanoparticles on the simulated porous media at depths.



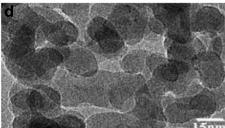


Figure 7 TEM bright field images of pure oxides (c) TiO₂ and (d) fumed SiO₂.

bending tension has no significant increase when bending rate increased, and in the end, its amount is lower than the basic fluid. On the other hand, by adding sodium hydroxide to basic fluid, bending tension increase regarding tension rate is much lower than its basic state. In fact, before two curves reach each other, at a fixed bending rate, mud has nanofumed silica and higher viscosity comparing to its basic state, while after increasing bending rate, the state changed for the fluids.

Like other industrial projects, feasibility of the samples are examined initially, then the results can be applied in field studies. Among the applications of this study is improvement in oil recovery which can be applied in many water wet reservoirs dominated by inhibition mechanism to extract more flow through really small caliber pores.

Conclusions

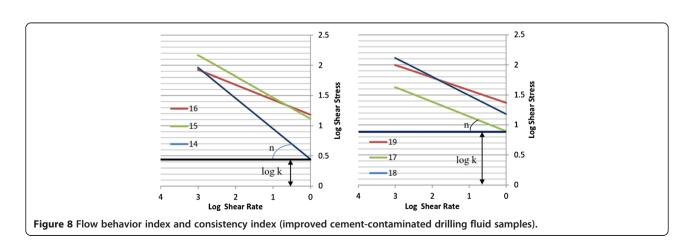
Using nanoparticles in all samples have resulted in recovery increase (Figures 4, 5, 6, 7). In all samples of flow with titanium dioxide nanoparticle, recovery is better except fluids with polyrhachioides and basic fluid, among which samples with fumed silica nanoparticles have the most recovery (Figure 8). Maximum recovery belongs to flows with sulfate and nano TiO₂. Considering the experiments, it is demonstrated that flows with nanoparticles

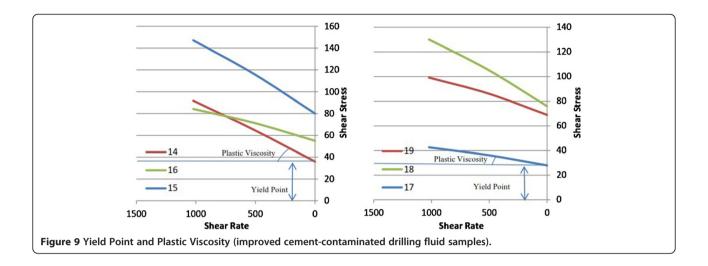
and, in particular, the titanium dioxide nanoparticles have the highest amount of recovery and thus using nanoparticles in water flooding projects and even some of the polymer flooding ones (for IOR process).

In laminar flow, increasing particles motion tendency toward the lower velocity area and consequently their displacement needing YP increase is very significant; too much increase in YP in drilling fluids with TiO_2 and fumed silica nanoparticles is among positive points of adding the matters to basic fluids obviously seen in diagrams covering laminar flows problem in retrospective ring spaces greatly (Figures 9, 10).

Among all fluids under study, fluids with TiO₂ nanoparticles have more YP in both states compared to fluids with fumed silica nanoparticles, and in sum, YP was higher in TiO₂ nano-fluid.

It is observed that with increasing sodium hydroxide, numerical amount of ${\bf n}$ for ${\rm TiO_2}$ nanoparticles drilling mud becomes higher than the other two. In sum, adding sodium hydroxide to the mud, ${\bf n}$ index decreases with a fixed, almost equal difference in nanoflows, and also maximum difference is seen in basic mud. The presence of fumed silica nanoparticles in particular with sodium hydroxide will have the minimum ${\bf n}$. Increasing sodium hydroxide, ${\bf k}$ will also increase and in the mean time, fumed silica nanoparticles and sodium hydroxide flow





will have higher n. Accordingly, it can cover low $\bf n$ in the flow (Figures 8). Additionally, although $\bf n$ is lower in fumed silica nanoparticle flows compared to flows with ${\rm TiO_2}$ nanoparticles and sodium hydroxide, coordination index is equal. Using nanoparticles in drilling mud will result in coordination index increase in all samples (Figure 9).

Treating cement contamination of drilling flow by sodium hydroxide (Figure 10), it is observed that flows composed of fumed silica nanoparticles have minimum **n**, while when the particles are in cement-contaminated mud, numerical value of **n** equals the **n** value in flow treated by sodium hydroxide without any nanoparticles. In other words, it is possible to treat the n value of cement-contaminated mud by fumed silica nanoparticles as effective as with sodium hydroxide. The flow with fumed silica nanoparticles and sodium hydroxide has minimum \mathbf{n} and maximum \mathbf{k} and maximum well-cleaning capability and results in high drilling rate by reducing viscosity at drill place.

Methods

In the first step, a certain amount of water is added to the compound and divided into eight plates. The first sample is considered as the base and left without change. To the second, fifth, eighth and eleventh samples, a certain amount of titanium dioxide is added and placed in the mixer. And fumed silica matter is added to samples 3, 6, 9 and 12 and then mixed to the extent that the mixture is homogenous. Then, each of the samples is separately located in pre-simulated environment. In 600s, the flow recovered from the device is registered in 20s intervals.



Figure 10 Effect of TiO₂ and fumed silica nano particles on the simulated drilling layers (drilling mud cake).

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

GC has contributed to all experiments and has been supervised by MH who supervised the project and also he is corresponding author. GC has participated in the statistical analysis and in the preparation of the manuscript. All authors read and approved the final manuscript.

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