

# A Critical Review of Wettability Alteration of Retrograde Gas Condensate Reservoirs Using Nanoparticles

## Abstract

Condensate and liquid blockage is a serious problem in gas condensate reservoirs as it reduces gas production. There are many methods to solve this problem, however, most of them are temporary or expensive. Wettability alteration of reservoir rocks from a liquid-wet state to a gas-wet state via nanoparticles is a long-lasting, cheap, and environmentally friendly solution to condensate blockage. With the aim of promoting this treatment in field scales, this review article presents a report of almost all the research carried out in this area. The results of different research teams are compared and the advantages and disadvantages of each research are detailed. Furthermore, the mechanisms and effects of gas-wetting alteration are fully explained, and the existence of an optimum wettability state is discussed. We found that silica nanoparticles are the most commonly used type of nanoparticles in wettability alteration towards a gas wet state due to their effectiveness and endurance. Most importantly, we present two new theories about the application of nanoparticles in the wettability alteration process of condensate reservoirs. First, it may be possible to inject nanoparticles into reservoirs via foam which not only stabilizes foam but also increases the effectiveness of wettability alteration treatment. Second, nanoparticles can be used to alter the wettability and prevent fines migration and sand production simultaneously. This review can be utilized as a reference in expanding the use of nanoparticles in gas-wetting alteration in field scales.

**Keywords:** Wettability alteration; Nanoparticles; Gas-wetting alteration; Sand production; Fines migration; Liquid blockage

## 1. Introduction

In retrograde gas reservoirs, condensate blockage can occur when the pressure is reduced to smaller than the dew point pressure because of production from the reservoir. Field examples of this phenomenon can be found in the literature (e.g., Briones et al.,<sup>[1]</sup> Hamoud et al.,<sup>[2]</sup> Bander et al.,<sup>[3]</sup> and many more in reference [4]). Condensate blockage is considered a challenge in the gas

production process since it substantially decreases gas relative permeability, and even low amounts of condensate can overwhelmingly slash gas production. Retained liquids (drilling and completion fluids, injected water, and connate water) can further intensify condensate/liquid blockage.<sup>[5]</sup> Moreover, liquid saturation can snowball to 70% in close vicinity of the wellbore,<sup>[6]</sup> and the experiments have shown that gas relative permeability will be reduced to zero if this saturation rises to 80%.<sup>[7]</sup> What is more, having calorific values higher than crude oil,<sup>[8]</sup> and being abundant and cheap to produce, natural gas has ever-growing importance for the world.<sup>[9,10]</sup> Considering all these along with the fact that a significant quantity of gas reservoirs are in the category of gas condensate reservoirs<sup>[11-13]</sup> means that condensate blockage is not a problem that can be neglected.

To tackle this problem, various methods have been proposed by researchers including water injection,<sup>[14]</sup> gas injection (utilizing methane, nitrogen, and carbon dioxide- mixtures of methane and nitrogen- and mixtures of methane, ethane, and propane),<sup>[15]</sup> solvent injection (methanol or dimethyl ether),<sup>[16]</sup> the use of energized fluids (supercritical CO<sub>2</sub>-foams),<sup>[17]</sup> drilling horizontal wells,<sup>[18]</sup> hydraulic fracturing,<sup>[19]</sup> acid stimulation,<sup>[20,21]</sup> inhibited diesel,<sup>[22]</sup> thermochemical treatments,<sup>[23]</sup> emitting microwave and ultrasonic waves,<sup>[24,25]</sup> and wettability alteration. All aforementioned methods tend to mitigate condensate build-up by elevating both gas and condensate production, and some of them can be implemented together. In a simulation-based process, Polat and Eren revealed that compared to mere horizontal wells, a combination of dry gas injection and horizontal wells leads to a higher condensate recovery factor and less condensate saturation around the wellbore.<sup>[26]</sup> The combination of hydraulic fracturing and wettability alteration treatments in a horizontal well located in North Texas led to a minimum leap of 300% in gas recovery.<sup>[27]</sup>

The wettability of a surface represents its tendency to be in contact with a certain fluid in the presence of other fluids.<sup>[28]</sup> Since two decades ago,<sup>[29]</sup> wettability enhancement from liquid-wet to liquid-repellent or gas-wet has been proposed as a means of alleviating condensate build-up in gas condensate reservoirs. Because reservoir rocks are liquid-wet, capillary forces are high; consequently, formed condensate and water in gas reservoirs imbibe into rock pores and spread over rocks' surfaces which leads to liquid blockage. Altering the wettability from liquid-wet to gas-wet state decreases capillary forces and the interfacial tension (IFT) between reservoir rocks and formed liquid. Thus, less water and condensate are imbibed to rock pores. Furthermore, the liquid formed in the vicinity of wellbores will be moved toward production wells as a result of the high-velocity gas flow and drag force,<sup>[30]</sup> and eventually, the liquid blockage will **be eliminated**. Wettability alteration of gas reservoirs to a gas-wetting state can be performed by using supercritical gases,<sup>[31]</sup> fluorinated surfactants, fluorinated polymers, fluorinated polymeric surfactants, and (fluorinated) nanoparticles.<sup>[32]</sup> Other techniques involving nonfluorinated silylation reagents carried in supercritical CO<sub>2</sub> have also been proposed by researchers.<sup>[33]</sup> However, their effectiveness in the presence of condensate has not been investigated yet.

As a consequence of surface and quantum effects, compared with their original size, shrinking the size of materials to a nanoscopic scale (less than 100 nanometers in one or more dimensions)

results in upgraded mechanical, thermal, magnetic, electronic, optical, and catalytic characteristics.<sup>[34]</sup> This justifies the broad usage of nanomaterials in various fields. Generally, nanomaterials and nanoparticles are employed in drugs and medications, manufacturing, environment, electronics, energy harvesting, and mechanical industries.<sup>[35]</sup> Only in petroleum engineering, nanomaterials and nanoparticles are utilized in self-dissolving frac plugs, sensors, composites, antimicrobial agents, catalysts, lubricants, coatings (both upstream and downstream sections), formation damage alleviation, enhancing in-situ and on-situ oil mobility, enhancing oil recovery, drilling fluids, and foam stabilizers.<sup>[36-40]</sup> More cases of the current and possible future applications of nanomaterials in the oil and gas industry can be found in Lau et al.<sup>[41]</sup>

This paper sifts through the existing literature regarding the wettability alteration of retrograde gas condensate reservoirs via nanoparticles, which alleviates condensate build-up around the wellbore. Different research works are compared and the flaws and advantages of each research are highlighted. The goal of this review article is to provide other researchers and experts in this field with a general description of almost all the research that has been conducted in this area, and eventually to promote the wettability enhancement of gas condensate reservoirs by nanoparticles as a common procedure in the field scale. The authors believe that this is plausible due to the following three convincing reasons:

- a) Required nanoparticles for this treatment are cheap, abundant, and environmentally friendly, meaning that there are no limitations in deploying high quantities of them.
- b) In general, wettability alteration treatments are cheaper than other methods such as drilling horizontal wells and hydraulic fracturing. Moreover, in the long run, the fact that this treatment is of a permanent nature makes it less costly than other temporary methods that need re-implementation.<sup>[42]</sup>
- c) Both the number and diversity of materials and methods used in published laboratory work on wettability alteration of gas reservoirs using nanoparticles have reached a notable figure, most of which are reported in this paper. Hence, it is time to expand pilot experiments and ultimately implement field-scale treatments.

## 2. Literature review

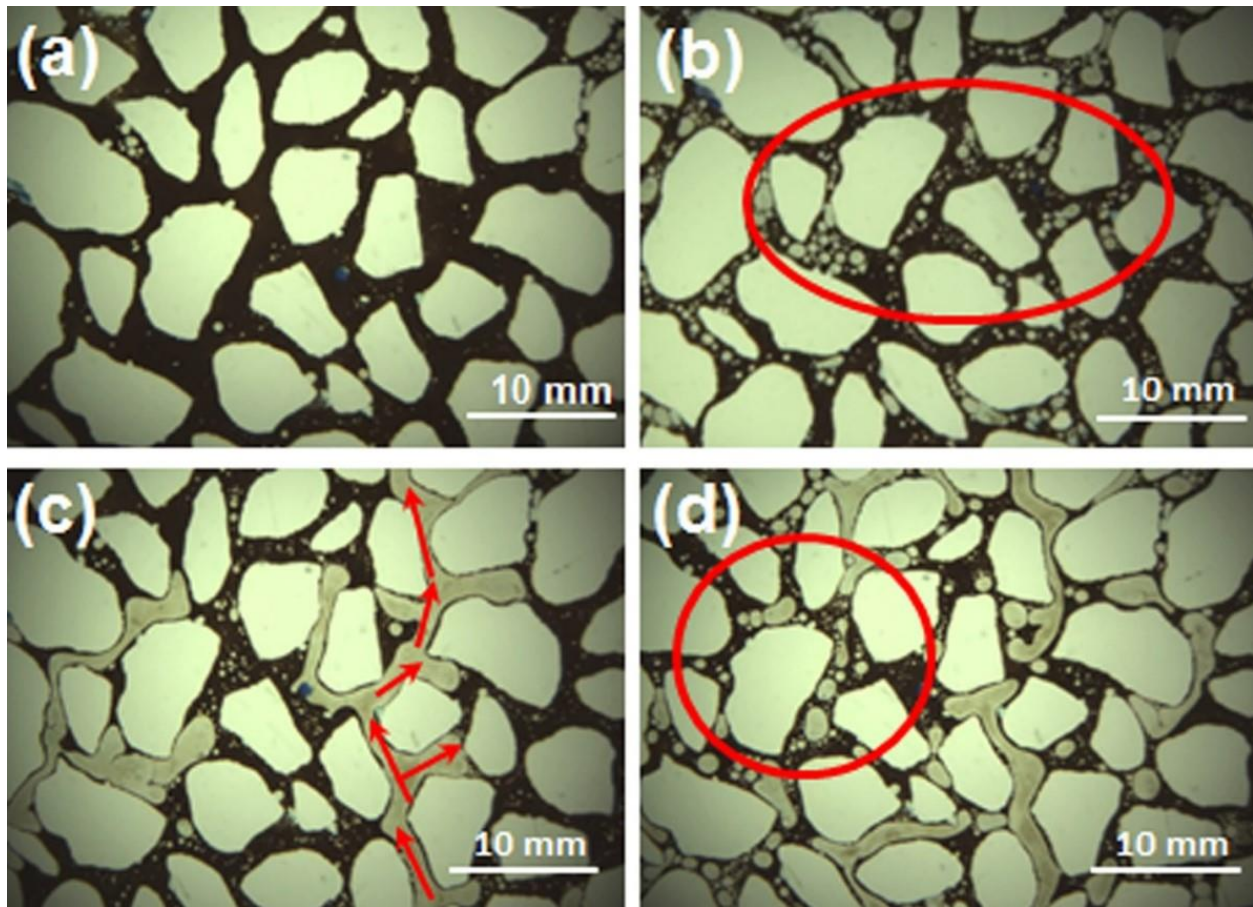
### 2.1. Silica nanoparticles (SiO<sub>2</sub> NPs)

By scrutinizing the literature, it seems that Mousavi et al.<sup>[43]</sup> were the first researchers **who** directly investigated the possibility of applying nanoparticles on retrograde gas condensate reservoirs to attain better wettability to solve the problem of condensate blockage. Any surface's wettability is in close relation to its roughness and its surface energy.<sup>[44]</sup> Taking this axiom into consideration, Mousavi et al. synthesized four groups of fluorinated silica nanoparticles (F-SiO<sub>2</sub> NPs). Fluorinating silica nanoparticles not only lessen surface free energy,<sup>[45]</sup> but also enhance their

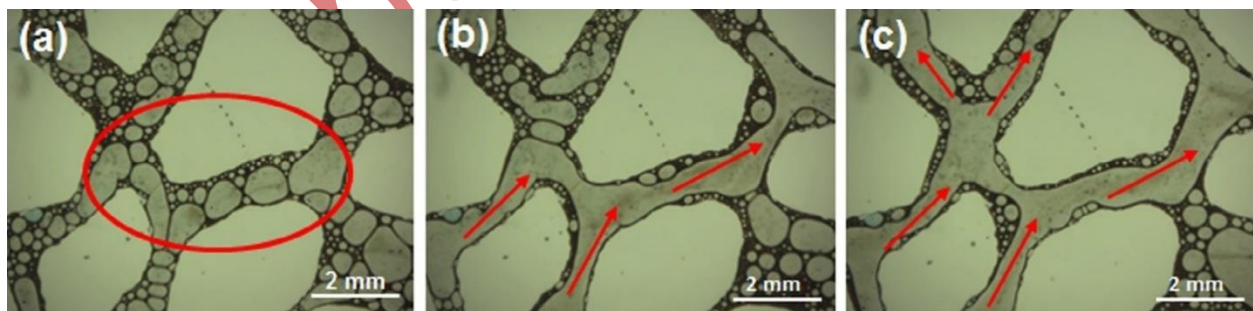
ability to form rougher surfaces;<sup>[46]</sup> Any substance capable of lowering surface free energy and roughening surfaces is apt to promote a gas-wetting state. The aforementioned groups of F-SiO<sub>2</sub> NPs were created via co-hydrolysis of tetraethylorthosilicate (TEOS) and a fluorinate alkylsilane (FAS) in ethanol; The usage of a fluoroalkylsilane would act as an oil/water repellent agent with notable thermal resistance properties in the solution. That is to say, the nanoparticles were durable and were not susceptible to unwanted reactions, which made them a reasonable option for usage in reservoirs. The Mentioned groups differed in FAS/TEOS ratios, 1:20 1:10 1:5 and 2:5, which led to the production of nanoparticles with diverse sizes. Limestone cores extracted from the Sarkhun reservoir in Iran were treated by generated F-SiO<sub>2</sub> NPs. While the formation of water and n-decane droplets on the samples' surface was not feasible before the treatment, contact angles of 147° and 61° for water and n-decane were achieved respectively after the treatment. Contact angle and imbibition tests are the most common tests for determining surface wettability. There are other methods of evaluating surface wettability. For example, the USBM-amott method, which is more concise than previous methods, is one of them. However, researchers seldom conduct this test as it is time-consuming.<sup>[47]</sup> Mousavi et al. conducted coreflooding tests to assess the efficiency of the treatment, and a 30% modification in pressure drop was reported after the wettability alteration from highly liquid-wet to intermediate gas-wet state. They considered the 1:5 ratio for FAS/TEOS as the optimum ratio as measured contact angles were all less in other ratios. Based on this, it was mentioned that the ratio should neither be too low not to decrease the surface's free energy, nor too high to decline surface roughness.

In most NP-based gas-wetting alteration treatments, NPs are added to roughen the surface. If the size of NPs is greater than an optimum amount, the surface roughness decreases. Hill et al.<sup>[48]</sup> showed that 50 nm Al<sub>2</sub>O<sub>3</sub> NPs form rougher surfaces than 135 nm ones. In the pore scale, the rougher the surface, the lower the capillarity of liquid bridges;<sup>[49]</sup> Low capillary pressure means that condensate accumulated in oil-wet pores displaces with more ease.<sup>[50]</sup> To reach super gas wettability on surfaces, Jin et al.<sup>[51]</sup> modified SiO<sub>2</sub> NPs with an average size of 40 nm by FG40, which led to a considerable reduction in the possibility of nanoparticles' aggregation apart from improving the efficiency of the solution. They succeeded in modifying a surface to a super gas-wet state by rising brine and n-decane contact angles from 23° and 0° to 152° and 127° using described fluorosurfactant-modified nano-silica. In addition to tests mentioned in previous researchers' work, Jin et al. utilized capillary tube rise tests to determine contact angles, however, this approach lacked application when the liquid level was lower than -22 mm in the capillary. Jin et al.<sup>[45]</sup> vividly demonstrated that in terms of wettability alteration, their modified NPs are superior compared to some fluorosurfactants and fluoropolymers in equal concentrations. In an ensuing effort, Jin et al.<sup>[52]</sup> illustrated via micromodeling **and visualized gas-flooding** that when both oil and gas phases are present within gas-wet pores, gas bubbles are apt to adhere to throat walls. As a result, oil will be departed with less hindrance. That is, achieving a gas-wet state counters liquid blockage and significantly decreases oil saturation. **This is vividly shown in Figs. 1-2.**





**Fig. 1** Visualized behavior of oil in the model in (a) the model saturated with crude oil; Oil saturation= 100%, (b) the model under gas-flooding; Oil saturation= 55/02%, (c) gas-wetting alteration of the model; Oil saturation= 42/76%, and (d) the end of gas-flooding; Oil saturation= 29/12%.<sup>[52]</sup>



**Fig. 2** Visualized behavior of oil in a gas-wet model in the presence of gas phase: (a) generated bubbles in a critical state of rupture, (b) bubbles tend to adsorb on the pore wall and a partial new path is created, (c) extrusion of oil based on the interaction between bubbles as they adsorb on the gas-wet walls.<sup>[52]</sup>

Saboori et al.<sup>[53]</sup> implemented a new nanofluid formulation to alter the wetting preference of carbonate retrograde gas condensate reservoirs. The nanofluid was synthesized by doping fluorine with silica and adding a coating of fluorosilane (F-SiO<sub>2</sub>-F). Synthesized NPs were semispherical in shape with a mean size of 50 nm. After applying the nanofluid, the authors reported a transition from liquid-wet to a highly gas-wet state based on achieved data of contact angle, surface energy, and fluid imbibition. The obtained wetting state was fixed for temperatures up to 150°C, and times more than 2 and 48 hours for water and oil, respectively. This thermal resistivity is of significance as it is higher than those reported by scholars who only applied chemicals and did not use NPs in their treatments.<sup>[54,55]</sup> If only the water contact angle is considered as a yardstick, a super gas-wet surface formed by fluoropolymer-modified NPs may not lose more than 7.5% and 13% of its wettability as a result of a 48-hour exposure to heat (150°C) and 45 minutes of ultrasonic wash, respectively.<sup>[56]</sup> Saboori et al. also demonstrated that F-SiO<sub>2</sub>-F nanofluid has a higher impact on rock's wettability than mere SiO<sub>2</sub> nanofluid.

Using F-SiO<sub>2</sub> NPs, Sayed et al.<sup>[57,6]</sup> were able to change the wetting state from strongly liquid wet to liquid repellent in sandstone. However, results from coreflooding experiments showed that in increasing relative permeability and decreasing pressure drop, synthesized NPs were inferior to a fluorinated polymeric surfactant used by Al-Yami et al.<sup>[58]</sup> What's more important is that the concentration of used nanoparticles should not have exceeded 0.35 wt% due to the possibility of agglomeration and pore throat blockage. But it is worth noting that just three pore volumes (PVs) of 0.065 wt.% concentration NPs caused a 40% rise in oil and gas relative permeabilities. Besides, coreflooding experiments revealed that despite the mentioned drawbacks, the coating of F-SiO<sub>2</sub> NPs on pores did not deteriorate even after 250 PVs of condensate fluids were flowed through the cores. In a treated reservoir, this means that in the long run, NPs will not detach from the pores during production, so the gas-wet state will be preserved.

Sakhaei et al.<sup>[59]</sup> enhanced the wettability of carbonate rocks to an intermediate gas-wet state by using F-SiO<sub>2</sub> NPs. What is interesting about their experiment is that contact angles of all tested liquids on treated samples moved upward after the treatment temperature was uplifted from ambient condition to 80°C. This was demonstrative of a second wettability enhancement and better effectiveness of F-SiO<sub>2</sub> NPs after the rise of temperature. The post-treatment increased contact angles after temperature rise were as follows: N-decane from 114° to 130°, n-hexadecane from 85° to 88°C, oil from 85° to 98°, condensate from 76° to 90°, disulfide oil from 62° to 80°, and brine from 113° to 126°.

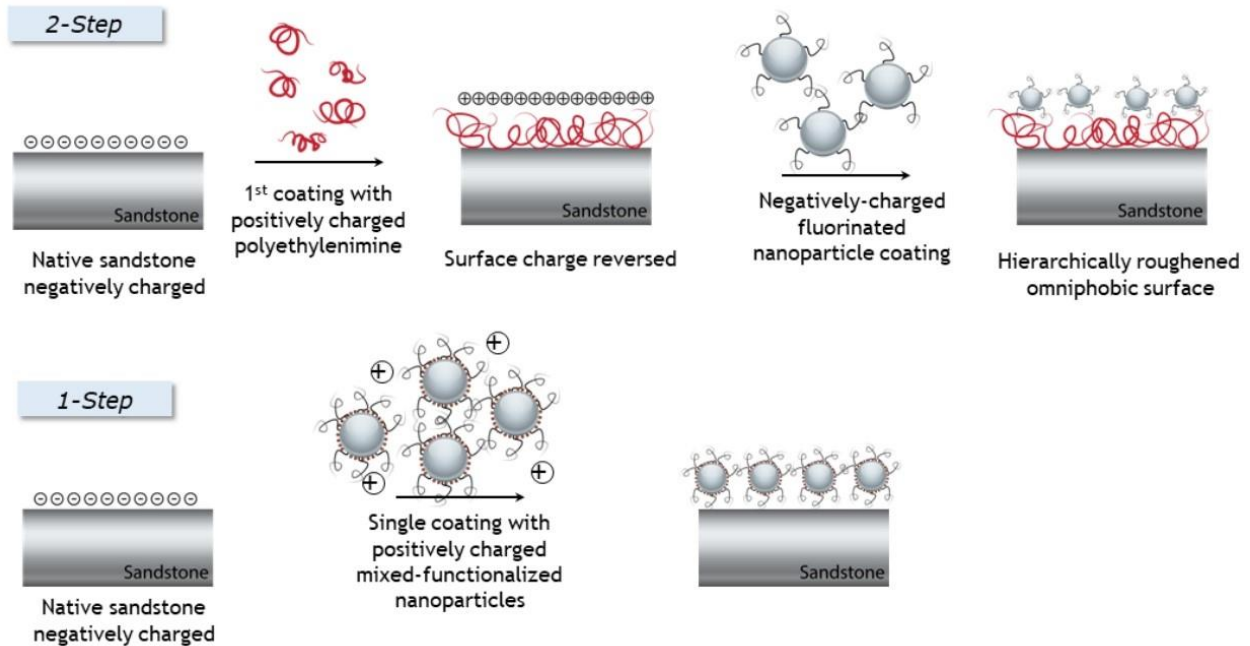
Gas reservoirs possessing very low matrix permeability are referred to as tight-gas reservoirs.<sup>[60]</sup> Franco-Aguirre et al.<sup>[61]</sup> were one of the first research teams who evaluated the performance of nanofluids in modifying the wettability of tight-gas reservoirs to a gas-wet state. First, by utilizing contact angle tests, they observed the effectiveness of different proportions of only Silnyl ®FSJ (SY- an anionic surfactant) to determine the best concentration for its solution, which was shown to be 0.4600 wt%. Second, they performed the same procedure on SiO<sub>2</sub> NPs and SiO<sub>2</sub> NPs modified with 3.0, 5.0, and 7.0 wt% SY in equal concentrations and it was discovered that 5 wt%

was the best amount for the named modifier. Subsequently, the effect of several concentrations of 5 wt% SY-modified SiO<sub>2</sub> NPs on contact angles was evaluated. Finally, a solution of 500 mg/L of 5 wt% SY-modified SiO<sub>2</sub> NPs with 0.46 wt% solution of SY was declared as the most desired nanofluid in terms of effectiveness and expense. Accordingly, the researchers chose this nanofluid for spontaneous imbibition tests and core-flooding tests in reservoir pressure and temperature. Rock samples used by this research team included both water-wet and oil-wet sandstones. All mentioned tests proved the alteration of wettability from liquid-wet to gas-wet for both types of samples. Furthermore, core displacement tests revealed that the permeability of all liquids improved after the treatment, and the team justified the gas permeability increase by linking it to the decrease in residual oil saturation ( $S_{or}$ ) values and subsequent rise in existing pore space.

Naghizadeh et al.<sup>[62]</sup> carried out another similar study using fluorine-doped SiO<sub>2</sub> NPs to reach a gas-wet state in carbonate reservoirs. What is distinct about their research is that they were the first to take into account the proportion of carbonate rocks' minerals (particularly calcite and dolomite) as a factor that affects the nanofluid treatment. The applicability of NPs in carbonate reservoirs is crucial because not only do carbonate reservoirs contain the greatest part of oil and gas sources but also the major portion of the world's hydrocarbon is extracted from them.<sup>[63,64]</sup> Based on their observations, it would only take five minutes for the nanofluid to alter the wettability to a stable gas-wet state. After imbibition tests, reduction factors ranging from 0.0016 to 0.773 for both brine and n-decane were reported, proving the alteration of wettability along with other tests. Naghizadeh et al. stated that the adsorption of nanoparticles is higher on more porous rocks. Via energy dispersive x-ray spectroscopy (EDX) analysis, the team uncovered that more Si and F elements get attached to carbonate rocks that are of calcite nature than those that are of dolomite. This was believed to be due to the surpassing reactivity of calcite compared to dolomite. Hence, more variance was reported for contact angles of calcite samples after surface modification. And generally, better recovery rates were witnessed for samples that had more calcite, demonstrating that F-SiO<sub>2</sub> NPs were more efficient on limestone rocks.

Sayed et al.<sup>[65]</sup> indicated that SiO<sub>2</sub> NPs can be modified more than once. They developed two-functionalized SiO<sub>2</sub> NPs. They noted that **unless the rock's surface is pre-treated with charged coatings**, F-SiO<sub>2</sub> NPs may not ruggedly and evenly get attached to some surfaces, particularly surfaces with a negative charge (sandstone as an example) for two reasons: a) F-SiO<sub>2</sub> NPs' low surface energy, and b) F-SiO<sub>2</sub> NPs' negative surface charge. Hence, they justified the need for coupling agents that could functionalize SiO<sub>2</sub> NPs and facilitate strong bonds between SiO<sub>2</sub> NPs and rocks' surface. This implied that silica nanoparticles had to be modified twice which was shown by the term "two-functionalized silica nanoparticles". **As demonstrated in Fig. 3, functionalizing NPs in this way would eliminate the need to pre-treat the rock's surface.**



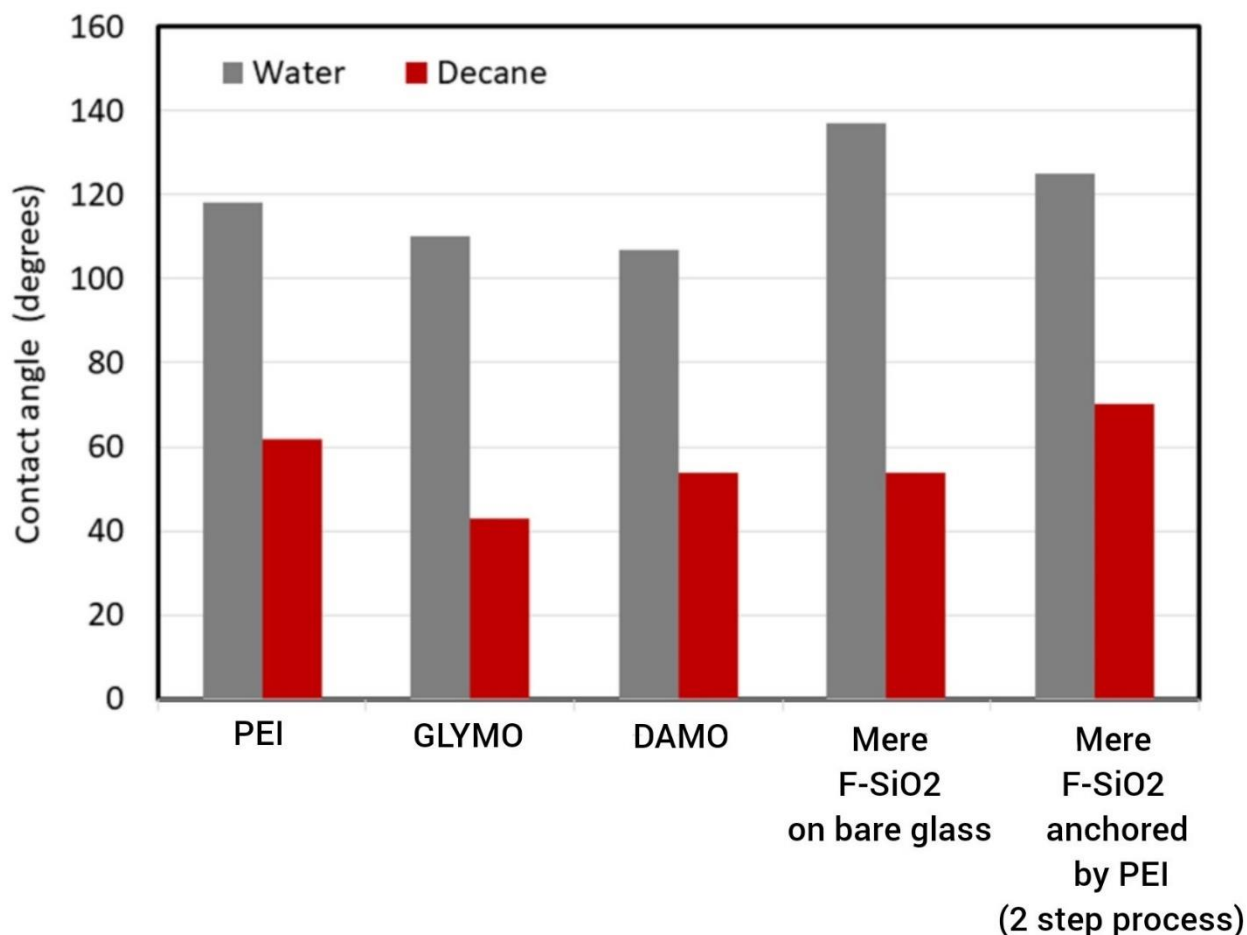


**Fig. 3** A summary of the two-step and one-step processes of covering rock's surface with NP coatings.<sup>[65]</sup>

To acquire two-functionalized NPs, Sayed et al.<sup>[65]</sup> functionalized SiO<sub>2</sub> NPs with fluoro-silane and a positively charged coupling agent. They investigated the effectiveness of polyethylenimine (PEI), N-(2-Aminoethyl)-3-aminopropyltrimethoxysilane (DAMO-), and (3-Glycidoxypropyl) trimethoxysilane (GLYMO) as three different coupling agents that functionalized SiO<sub>2</sub> NPs before or after functionalization of SiO<sub>2</sub> NPs with fluoro-silane. Among the three coupling substances, the research team dismissed GLYMO because of its gelling property, and PEI was demonstrated to achieve the highest water and n-decane contact angles. The performance of different two-functionalized SiO<sub>2</sub> NPs is exhibited in Fig. 4. The team also uncovered that the performance of two-functionalized silica nanoparticles was influenced by the order of SiO<sub>2</sub> NPs' enhancement, which was fluorinating nanoparticles first and then modifying them with a coupling agent or vice versa. In comparison with treating samples with mere F-SiO<sub>2</sub> NPs, it was concluded that n-decane mobility soared as a result of treatment by two-functionalized SiO<sub>2</sub> NPs since the angles from which n-decane droplets would begin rolling off the samples were drastically reduced. Roll-off tests were also indicative of the superiority of double-functionalized SiO<sub>2</sub> NPs to F-SiO<sub>2</sub> NPs in resistance against salinity and decane. Noteworthy enough, the coreflooding test resulted in a 37.4% slash of pressure drop and 1.57 growth factor of gas relative permeability after only 4.5 PVs of two-functionalized SiO<sub>2</sub> NPs' injection, while obtaining similar results (a 40% plummet in pressure drop and an increase factor of 1.69 for gas relative permeability) via a fluorinated polymeric surfactant required 15 PVs of injection. On the other hand, it was revealed that compared to F-SiO<sub>2</sub> NPs, double-functionalizing SiO<sub>2</sub> NPs would lead to a reduction in water and in some cases n-decane contact angles. For PEI, this reduction was about 18° for the water contact



angle, but an increase of  $8^\circ$  was observed for n-decane contact angle. However, better durability, higher decane mobility, and less treatment injection volume are needed, thus, a lower overall price regarding two-functionalized  $\text{SiO}_2$  NPs overshadows the reduction in water contact angles.



**Fig. 4** Ambient condition contact angles of water and n-decane on glass treated with different two-functionalized  $\text{SiO}_2$  NPs.<sup>[65]</sup>

Ganie et al.<sup>[66]</sup> utilized Perfluorooctanoic acid and perfluorononanoic acid as two new elements to modify  $\text{SiO}_2$  NPs. Although the coating process was successful, their work lacked a direct assessment of formed fluoroalkanoic acid-modified NPs' effectiveness on wettability alteration toward gas wetting in gas-condensate reservoirs.  $\text{SiO}_2$  NPs were also employed by other researchers whose works are reported further in this article. Owing to the growing improvement and advantages of  $\text{SiO}_2$  NPs, the use of them in gas-wetting alteration is still being subjected to research. Recently, Tu et al.<sup>[67]</sup> examined wettability change to super gas-wet conditions by modified  $\text{SiO}_2$  NPs and indicated that this condition will be preserved in both acidic and alkaline media.

Another recent application of  $\text{SiO}_2$  NPs is in sand production prevention. It has been shown that  $\text{SiO}_2$  NPs can coat sand grains and hinder sand migration/production.<sup>[68,69]</sup> The applied

nanoparticles should not widely reduce permeability.<sup>[70]</sup> This method is very recent and the number of literature in this regard is a handful. Moreover, the applicability of other types of NPs in preventing sand production needs to be confirmed.

Regarding health, safety, and environment (HSE), the leakage of chemicals of reservoir treatment into the soil, underground waters, and subsequent hazards for life forms has always been of concern. While some researchers may consider SiO<sub>2</sub> NPs harmless,<sup>[71]</sup> others may categorize them as toxic.<sup>[72]</sup> The toxicity and negative impacts of SiO<sub>2</sub> NPs on the environment are very case-dependent generally. They are in direct connection with their concentration, size,<sup>[73]</sup> synthesis method,<sup>[74,75]</sup> and exposure type (dietary, respiratory, cellular scale, etc.). Moreover, in some cases, SiO<sub>2</sub> NPs may act as harmless materials that interact with other toxicants and intensify their toxicity.<sup>[76]</sup>

## 2.2. Metal oxide nanoparticles

While metal oxide NPs are one of the most commonly used NPs in enhanced oil recovery (EOR),<sup>[77]</sup> they still have limited application in gas-wetting alteration in condensate reservoirs.

Esmailzadeh et al.<sup>[78]</sup> were the first who applied NPs to weaken condensate blockage by promoting a “super gas-wetting state”. In addition to using nano-silica, they proposed titanium oxide nanoparticles and multiwall carbon nanotubes (CNTs) to be used as wettability changer agents for carbonate rocks. They prepared TiO<sub>2</sub> and SiO<sub>2</sub> NPs via the sol-gel method, and subsequently made SiO<sub>2</sub>, TiO<sub>2</sub>, and CNT nanofluids which also included polytetrafluoroethylene (PTFE), 2,2,2-Trifluoroethanol (TFE), and Trichloro(1H,1H,2H,2H-perfluorooctyl)silane (PFOS) for further reduction of surface free energy. They eventually managed to change the water contact angles from 0° to 161°, 164°, and 163° for TiO<sub>2</sub>, SiO<sub>2</sub>, and CNT nanofluids. N-decane contact angles were also altered from 0° to 144°, 151°, and 147° for the above nanofluids. During these experiments, the hydrophilic state of the rock was changed and a super hydrophobic gas-wetting state was obtained. Moreover, water imbibition into rock was zeroed and the treatment was completely stable for temperatures up to 160°C.

Esmailzadeh and Sadeghi<sup>[79]</sup> synthesized ZnO and SiO<sub>2</sub> nanocomposites using the wet-chemical co-precipitation method. Subsequently, they combined PTFE, TFE, ethanol, and a mixture of PFOS with the aforementioned nanocomposites to structure a nanofluid that could affect carbonate reservoir rocks. The goal of using nanocomposites was to gain a rougher surface and listed fluorochemicals were added to lessen surface free energy as they had very low surface energy due to –CF<sub>3</sub> and –CF<sub>2</sub> bonds. Contact angle tests were conducted after washing the samples with toluene and methanol and treating them with the obtained nanofluid; while the samples were entirely wet to water and oil before the treatment, contact angles rose to 162° for water and brine and roughly 136° for n-decane and condensate, indicating an ultra gas-wet surface. Core displacement tests were carried out to simulate the nanofluid’s performance under the reservoir’s conditions, and a nearly 56% plunge in pressure drop for both gas/water and gas/oil flows was

reported after the treatment. Ultimately, not only was a state of super oil/water repellency achieved but also the surface gained self-cleaning properties. In terms of changing wettability, this method was claimed to be superior to previous chemical treatments.

Extending their previous research, Esmailzadeh et al.<sup>[80]</sup> evaluated the effectiveness of only TiO<sub>2</sub> NPs in optimizing wettability in reservoir conditions. By synthesizing superamphiphobic TiO<sub>2</sub> NPs whose mean diameter was 20-50 nm, they altered the wettability of strongly water-wet and oil-wet samples to super gas-wet. Using contact angle tests, they concluded that the performance of TiO<sub>2</sub> nanofluid is not limited by the high salinity of fluids and the primary wettability of rock. However, the substance would form insignificantly better oleophobicity in highly water-wet samples compared with highly oil-wet samples. Via testing the imbibition of the nanofluid into cores, the authors deduced that the efficiency decrease of the nanoparticles will be marginal in case of condensate saturation presence in rocks and the above-mentioned nanofluid is capable of imbibing into gas-condensate saturated rocks, subsequently changing the wettability, and eventually improving the gas-condensate recovery.

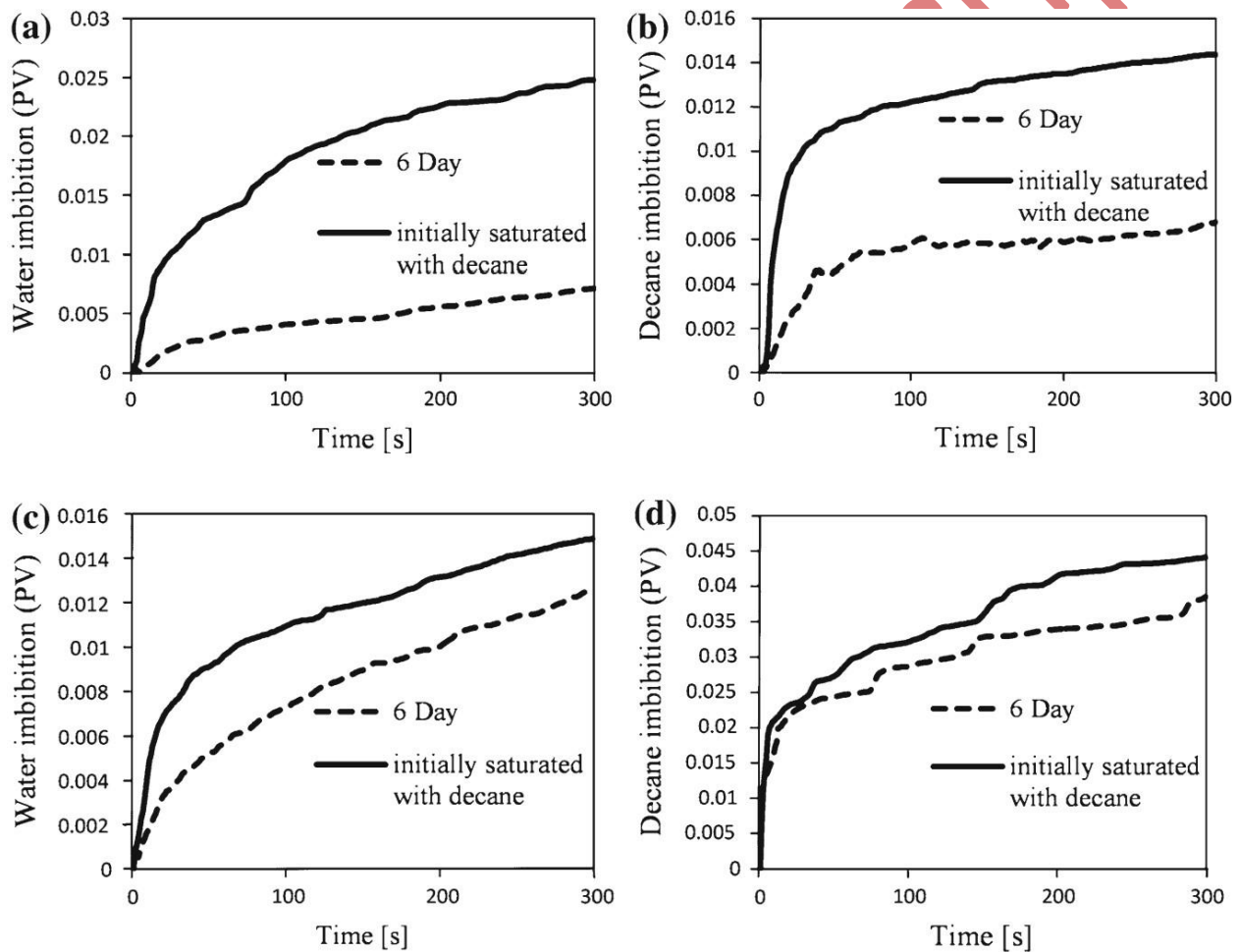
To enhance the wettability of carbonate condensate reservoirs to gas wetting, more new nanofluids were introduced by Safaei et al.<sup>[81]</sup> They developed two distinct nanostructures by coating Fe<sub>3</sub>O<sub>4</sub> NPs with Poly (PAV) and Hydroxyapatite (HAp). These nanoparticles are advantageous as in certain cases, they can be recovered from well fluids via magnetic fields. In total, Fe<sub>3</sub>O<sub>4</sub>-HAp NPs were proven to be more effective than Fe<sub>3</sub>O<sub>4</sub>-PVA in treating samples. Treating samples with hydroxyapatite-coated iron oxide NPs led to imbibition decreases of 48 and 75%, imbibition reductions of 85 and 64% in low permeability samples (less than 3 md), pressure drop reductions of 31 and 80%, and pressure drop reductions of 83 and 32% for water and gas-condensate in low permeability samples in the mentioned order. On top of that, by contact angle tests, the researchers demonstrated that the higher the temperature is (up to 80°C), the better wettability is altered. And the treatment was stable for pressures up to 40 bars.

More cases of using metal oxide NPs will be mentioned in section 2.5.

### **2.3. SurfaPore M**

Aminnaji et al.<sup>[82]</sup> were also among the pioneers in utilizing nanofluids for gas condensate reservoirs. SurfaPore M, a cost-effective environmentally-green water-based nanofluid consisting of silicon-based molecules and fluoro-polymer that was developed to create water and oil repellency on surfaces, was employed to modify the characteristics of carbonate and sandstone rocks whose permeability was 30 md and 190 md. Samples were drowned in pure SurfaPore M at 70°C for three days for contact angle and imbibition tests. Scanning electron microscope (SEM) images proved the presence of a thin layer of organofluorine and silicon-based NPs on surfaces after the treatment. A significant leap in contact angles was achieved after treating rocks with the aforementioned nanofluid. Moreover, the surface free energy of both rocks was reduced to just over 4 mN/m due to the existence of organofluorinated chemicals in the nanofluid. The imbibitions

of water and decane decreased about 96% and 79% for carbonate and 97% and 91% for sandstone samples. An advantage of Aminnaji et al.'s research is assessing the impact of initial decane saturation on the performance of nanofluid by imbibition tests. In the case of complete initial saturation with decane, the results were indicative of water imbibition reductions of 83.3% and 96.4%, and n-decane imbibition reductions of 55.7% and 89.6% for carbonate and sandstone samples, respectively (Fig. 5). Through core displacement results, it was calculated that the brine effective permeability at residual gas saturation jumped from 57 md before the treatment to 130 md after the treatment for the sandstone sample. In general, the rocks' wettability was successfully changed from liquid-wetting to intermediate gas-wetting based on every experiment conducted. Aminnaji et al. also showed that the presence of more initial saturation will result in less influence of nanofluid on surfaces.



**Fig. 5** Imbibition into treated rocks aged for 6 days saturated with air and treated rocks saturated with normal decane: (a) water imbibition into a carbonate rock, (b) n-decane imbibition into the carbonate rock, (c) water imbibition into a sandstone rock, (d) n-decane imbibition into the sandstone rock.<sup>[82]</sup>



Gahrooei and Ghazanfari<sup>[83]</sup> expanded the study of Aminnaji et al. by exploring SurfaPore M nanofluid and its applicability in changing sandstone reservoir rocks' wettability using various tests. By manipulating this nanofluid, they altered the wettability of sandstone reservoir rocks from strongly liquid-wet to intermediate gas-wet after 24 hours of aging time. This accomplishment was confirmed via corresponding results obtained from coreflooding, contact angle, and spontaneous imbibition tests, and the used nanofluid was resistant to brine salinities up to 130,000 ppm in ambient conditions. Additionally, to indicate the adsorption of SurfaPore M nanofluid on samples, adsorption tests were done in preliminary concentrations of 0.25, 0.5, 1, 1.5, and 2% w/w. The results manifested that regardless of the concentration, the amount of SurfaPore M nanofluid adsorbed on sandstone hardly increased after 10 hours. And the higher the concentration of NPs was, the higher the adsorption was. More importantly, they considered intraparticle diffusion and external mass transfer rate as two factors that essentially influence the adsorption rate. Shayesteh et al.<sup>[84]</sup> confirmed the findings of Gahrooei and Ghazanfari regarding the contribution of intraparticle diffusion to the adsorption rate of SurfaPore M. Besides, they emphasized on boundary layer diffusion mechanism as another factor that affects the adsorption rate of NPs.

By conducting coreflooding experiments, Gahrooei and Ghazanfari illustrated that SurfaPore M nanofluid is not workable in low-permeability sandstone reservoir rocks. This was stated to emanate either from pore blockage or clay swelling which led to a decline in permeability and a subsequent rise in pressure drop. Their research revealed that SurfaPore M nanofluid must not be used in low permeable and particularly clayey reservoirs.

To provide a solution, Gahrooei et al.<sup>[85,86]</sup> suggested that MariSeal 800, an industrial fluorinated chemical agent, can be used as an alternative to SurfaPore M in low permeability reservoirs as it would not lead to any considerable formation damage.

It is possible to (partially) counter clay swelling by injecting SurfaPore M in a foam-based carrier fluid since less water will be injected into the reservoir due to the nature of the foam. Also, foam as a carrier slashes gravity segregation and viscous fingering and increases the viability of displacement front and sweep efficiency.<sup>[87]</sup> However, we did not find any case of utilizing foam as a carrier fluid of NPs in the gas-wetting alteration of reservoirs.

Gahrooei and Ghazanfari mixed SurfaPore M nanofluid with different concentrations of NaCl brine and let them rest for a month to determine its resistivity against salinity via visual detection of nanoparticle precipitates. Tabar et al.<sup>[88]</sup> adopted a different approach to determine the stability of SurfaPore M. They exposed SurfaPore M-modified calcite samples to 0.5 %wt concentration of NaCl brine and MgCl<sub>2</sub> brine for two days. Subsequently, based on the brine type, cores' chosen area for contact angle tests (top or bottom), and exposed cores' situation (horizontal or vertical) reductions in water advancing contact angles ranging from 10-24° were recorded, however, no great reduction was recorded for kerosene and dead oil contact angles. It can be of great importance that MgCl<sub>2</sub> brine had a slightly more detrimental impact on the water advancing contact angles of all cores. This research team also proved that the deteriorating effect of salinity on contact angles can be wiped out by washing off the salt present in samples.

## 2.4. Other novel nanoparticles

Wang et al.<sup>[89,90]</sup> reported the successful application of ether surfactant nanofluid CNDAD1# for the wettability alteration of sandstone cores to a gas-wet state without increasing surface roughness. Nevertheless, their work targeted a tight gas sandstone reservoir in which the condensation phenomenon had not occurred, and the research aimed to alleviate the water locking problem originating from water build-up in the drilling or fracturing process. The effectiveness of CNDAD1# nanofluid was proven via contact angle, spontaneous imbibition, coreflooding, and dynamic and static adsorption experiments, and only brine was utilized for this purpose. Their results suggested that it is safe to argue that CNDAD1# nanofluid will also be effective in mitigating condensate blockage in retrograde gas reservoirs.

Ahmadi and co-workers<sup>[91]</sup> offered to use natural calcium carbonate nanoparticles (Bio-Ca NPs) as a wettability alteration agent and a remedy for condensate blockage in gas reservoirs. Sand pack experiments were conducted to simulate condensate recovery. The ability of Bio-Ca NPs to form gas-wet surfaces was presumed to be due to the presence of chitin in their biostructure. The formed NPs were harmless to the environment and were economical to use as they were created from natural sources. And they were not susceptible to agglomeration due to the adsorption of Cetyl trimethyl ammonium bromide (CTAB) molecules on them. The Bio-Ca NPs were utterly advantageous to SiO<sub>2</sub> NPs in surging condensate contact angles and reducing pressure drop. However, especially about condensate contact angles, the result may have been the opposite if modified SiO<sub>2</sub> NPs had been used instead of mere SiO<sub>2</sub> NPs. Bio-Ca NPs were subjected to further sand pack tests by Seifi et al.<sup>[92]</sup> In their tests, the cases in which Bio-Ca NPs were injected into the sand pack with lower flow rates showed better results than the higher flow rates of injection. This was attributed to the longer contact time of the Bio-Ca NPs and pores in lower flow rates of injection.

Shayan and Esmaeilnezhad<sup>[93]</sup> surged water and oil contact angles on carbonate rocks from 20° and 4° to 90° and 52° in sequence and changed the wettability from oil-wet state to gas-wet state

by the use of NFS-104, a water-based nanofluid which is considered to be environmentally-friendly since it does not contain fluorocarbon particles. The wettability alteration process and its efficiency were only evaluated via contact angle tests and no further experiments were performed.

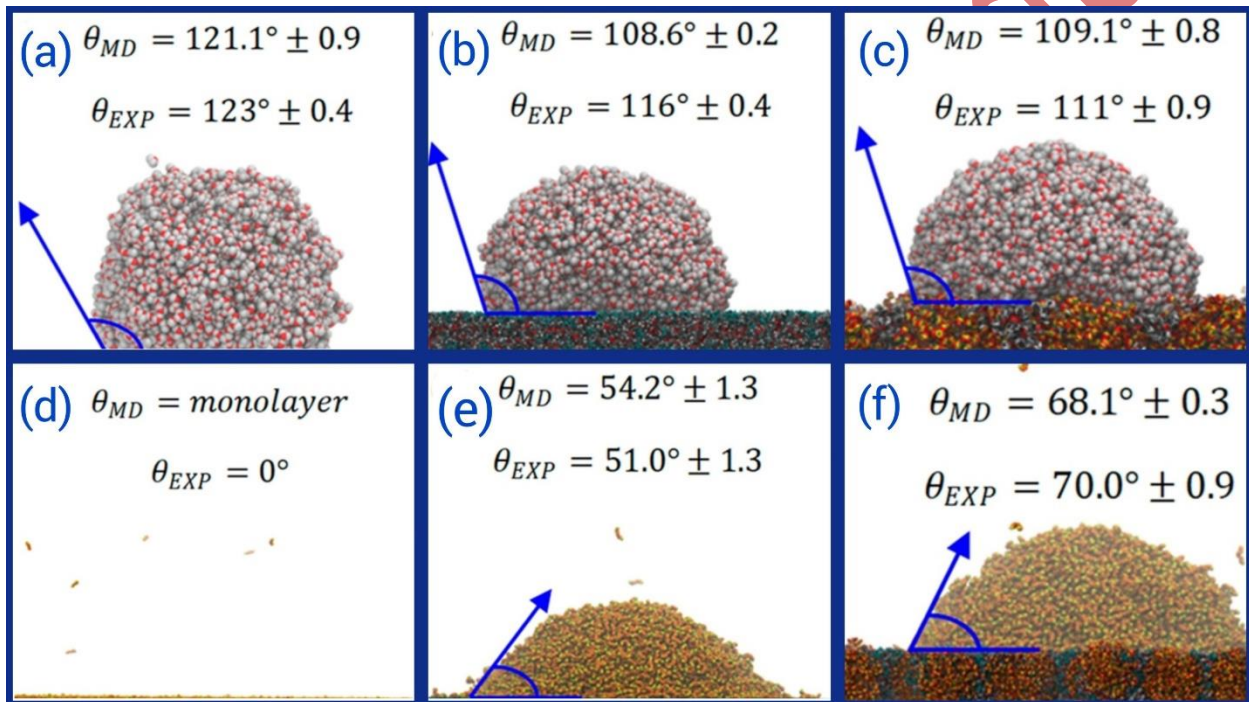
Wang et al.<sup>[94]</sup> fluorinated Nanocrystalline cellulose (NCC) by which they were able to change the wettability to super gas-wetting. The success of the fluorine modification process of NCC was demonstrated by x-ray photoelectron spectroscopy. The enhancement in surface wettability was made evident as surface free energy, capillary tube level, and imbibition and injection pressure of liquids (brine, n-decane, oil) into samples all considerably diminished while brine and n-decane contact angles soared. NCC can also be used to modify platinum, gold, silver, palladium, and nickel nanoparticles.<sup>[95]</sup> It is completely environmentally green and can be created from biomass and even biowaste,<sup>[96,97]</sup> which means that there are countless sources from which it can be attained. Moreover, by the use of a waste-free technology, the total cost of producing one kg DM of nanocrystalline aggregates will be only \$3.5-4.0.<sup>[98]</sup> These advantages turn this material into a suitable candidate for wettability alteration usage.

## 2.5. Numerical simulation-based research

Sepehrinia and Mohammadi<sup>[99]</sup> ran molecular dynamics (MD) simulations to assess the role of F-SiO<sub>2</sub> NPs in changing the wettability of silica nanopores in the presence of water or n-decane. They defined 7 nm as the size of pores and 3 nm as the diameter of NPs. They deduced that these NPs can form hydrophobic surfaces and slash static water blockage. However, they do not affect the presence of decane particles in pores. Their most noteworthy conclusion was that NPs needed more energy to be accumulated than to be absorbed on surfaces. This means that the possibility of formation damage via aggregation of nanoparticles is very low.

To gain a broader understanding of wettability alteration, Moncayo-Riascos et al.<sup>[100]</sup> combined the work of Sepehrinia and Mohammadi and Franco-Aguirre et al.; And using both experimental and MD simulation methods, they assessed the capability of SiO<sub>2</sub> NPs that were modified with 20 wt% of Silnyl FSJ surfactant in wettability enhancement of sandstones. Moncayo-Riascos et al. approached the experimental aspect of the research similar to previous literature. Furthermore, regarding MD simulations, they defined 10-4-3 wall potential and concise amounts of adhesion and cohesion forces and correlations between them to simulate a solid oil-wet surface. They utilized a consistent valence force field (CVFF)<sup>[101]</sup> to simulate SY surfactant. Moreover, the transferable potentials for the phase equilibria-united atom force field (TraPPE-UA),<sup>[102]</sup> simple point charge potential (SPC-E),<sup>[103]</sup> and CVFF models were applied to define n-decane and brine in simulations. Moreover, in light of the outcomes of experiments, Moncayo-Riascos and his research team managed to simulate SiO<sub>2</sub> NPs modification by SY surfactant and subsequently to model an initially oil-wet sandstone covered with SiO<sub>2</sub> NPs functionalized with Silnyl FSJ fluorocarbon surfactant. In addition to experimental contact angles, contact angles were modeled and calculated in MD simulations under equal conditions, which were perfectly consistent with contact angles measured via physical experiments, illustrating the accuracy of simulations. Both

experimental and simulational data were demonstrative of wettability change from oil-wet to gas-wet. In the end, by comparing brine and n-decane contact angles achieved from experimental and simulational data, the researchers deduced that NPs merely aid fluorocarbon surfactant in reaching a gas-wet state as the presence of NPs did not drastically affect contact angles; **This is clearly illustrated in Fig. 6.** However, based on simulations using SiO<sub>2</sub> NPs along with fluorocarbon surfactant would slash a minimum percentage of 52 in terms of the amount of surfactant needed to obtain a gas-wet surface, meaning that the proposed nanofluid is economically superior to mere fluorosurfactant solutions. Besides, they noticed that the interaction between NPs and liquids is a function of the density of SY-modified NPs that coat the surface, and the way NPs cover the surface is impacted by the arrangement of surfactant adsorption on nanoparticles.



**Fig. 6** Experimented and simulated brine and n-decane contact angles: (a) brine on an untreated sample, (b) brine on a sample treated with mere Silnyl FSJ surfactant, (c) brine on a sample treated with 0.3 wt% of SY-functionalized SiO<sub>2</sub> NPs, (d) n-decane on an untreated sample, (e) n-decane on a sample treated with mere Silnyl FSJ surfactant, (f) n-decane on a sample treated with 0.3 wt% of SY-functionalized SiO<sub>2</sub> NPs.<sup>[100]</sup> The untreated sample is considered oil-wet.

Silnyl FSJ surfactant was also employed by Villegas et al.<sup>[104]</sup> to modify  $\gamma$ -alumina and magnesia nanoparticles. Conducting both laboratory contact angle tests and LAMMPS MD simulations, they proved the wettability alteration of sandstone rock from liquid-wet to gas-wet as a result of surface treatment by  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> and MgO NPs functionalized with SY. CVFF, SPC-E, and the Buckingham-Coulomb potential<sup>105</sup> were employed to model the surfactant, water molecules, and NPs, respectively. Contact angle evaluations by both laboratory and MD simulation indicated that  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> NPs functionalized with 30% of Silnyl FSJ preceded MgO NPs modified with the same



concentration of Silnyl FSJ in forming liquid-repellent surfaces. Modelings were also demonstrative of slashes of 88% and 42% for water and n-decane adhesive energies and unchanged water and n-decane cohesive energies as a result of implementing 30 wt% Silnyl-modified  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> nanoparticles. This work was experimentally continued by Galeano-Caro et al.<sup>106</sup> who combined NP injection and gas injection methods. They were the first to use a flue gas stream as the carrier fluid for the two aforementioned functionalized NPs to reach a gas-wetting state. This method is of high applicability in tight gas reservoirs due to the minimum size of gas particles as an NP carrier. The method was proved to be successful by a 57% of reduction in residual oil saturation of tested outcrops.

A summary of the different investigations mentioned in this article is presented in Table 1. Contact angle, imbibition, and coreflooding are the three most commonly used tests to assess and confirm wettability alteration in these experiments. However, only the results of contact angle tests are gathered in table 1 due to two reasons: (a) imbibition and coreflooding tests were not carried out by all the researchers. (b) the results of imbibition and coreflooding tests are directly affected by the condition of the experiment and properties of used cores which both vary in each research.

**Table 1. A summary of different investigations reported in this paper**

Researchers	Used nanoparticles	Treated sample	Achieved water contact angles	Achieved decane contact angles
Mousavi et al. <sup>[43]</sup>	F-SiO <sub>2</sub>	Limestone	147°	61°
Jin et al. <sup>[45,51,52]</sup>	F-SiO <sub>2</sub>	Sandstone	152°	127°
Sayed et al. <sup>[6,57]</sup>	F-SiO <sub>2</sub>	Sandstone	148.5°	51°
Shakhaei et al. <sup>[59]</sup>	F-SiO <sub>2</sub>	Carbonate	126°	130°
Naghizadeh et al. <sup>[62]</sup>	F-SiO <sub>2</sub>	Carbonate	155°	113°
Esmailzadeh et al. <sup>[78]</sup>	Modified SiO <sub>2</sub>	Carbonate	164°	151°
Esmailzadeh & Sadeghi <sup>[79]</sup>	Modified SiO <sub>2</sub>	Carbonate	162°	136°
Franco-Aquirre et al. <sup>[61]</sup>	SY-Modified SiO <sub>2</sub>	Sandstone	118°	95°
Moncayo-Riascos et al. <sup>[100]</sup>	SY-Modified SiO <sub>2</sub>	Sandstone	111°	70°

Saboori et al. <sup>[53]</sup>	F-SiO <sub>2</sub> -F	Carbonate	146.5°	138° (oil)
Sayed et al. <sup>[65]</sup>	Two-Functionalized SiO <sub>2</sub>	Sandstone	118°	62°
Esmailzadeh et al. <sup>[78]</sup>	Modified TiO <sub>2</sub>	Carbonate	161°	144°
Esmailzadeh et al. <sup>[78]</sup>	Modified CNT	Carbonate	163°	147°
Esmailzadeh & Sadeghi <sup>[79]</sup>	Modified ZnO	Carbonate	162°	136°
Esmailzadeh et al. <sup>[80]</sup>	Modified TiO <sub>2</sub>	Carbonate	162°	143°*
Safaei et al. <sup>[81]</sup>	Modified Fe <sub>3</sub> O <sub>4</sub>	Carbonate	135°	110°*
Aminnaji et al. <sup>[82]</sup>	Surfapore M	Carbonate	139°	95°
Aminnaji et al. <sup>[82]</sup>	Surfapore M	Sandstone	138°	104°
Gahrooei & Ghazanfari <sup>[83]</sup>	Surfapore M	Sandstone	128°	85°
Shayesteh et al. <sup>[84]</sup>	Surfapore M	Carbonate	138°	73°
Ahmadi et al. <sup>[91]</sup>	Natural CaCO <sub>3</sub>	Carbonate	Not reported	105°*
Wang et al. <sup>[94]</sup>	Modified NCC	-	153°	142°
Pablo Villegas et al. <sup>[104]</sup>	SY-Modified $\gamma$ -Al <sub>2</sub> O <sub>3</sub>	Sandstone	134°	123°
Pablo Villegas et al. <sup>[104]</sup>	SY-Modified MgO	Sandstone	128°	114°

\*= Condensate contact angle

As can be observed from Table 1, wettability is changed to a neutral and or gas-wet state in all cases which is indicative of the capability of different NPs. In addition to different nanoparticles, modifiers, and concentrations used, variations of reported contact angles stem from differences in several other factors:

- (a) Rock type and properties of rocks used in tests.
- (b) The temperature at which contact angles are measured.

(c) Experiment procedure, which includes the time and temperature in which samples are drowned in nanofluid; And, whether the surface of the rock is polished before the droplets are placed on the rock.

(d) The contact angle changes with the size of the liquid droplet placed on the rock,<sup>[107]</sup> which affects the accuracy of measurements.

All the above-mentioned factors vary in experiments reported in this article.

### **3. The optimum wettability condition**

The final aim of every enhanced oil and gas recovery procedure is not to improve production, but it is to maximize production. Many research teams have confirmed that altering the affinity of rock from liquid-wet to gas-wet or super gas-wet state increases gas and condensate production. A matter that can be of question is the existence of a wettability state in which production amount reaches its peak. And in case of the existence of such a state, which state of wettability condition is best to be targeted in treatments?

In 2015, by utilizing the effect of capillary pressure and relative permeability on the contact angle, a mathematical model was proposed by Naik et al.<sup>[108]</sup> to solve the water blockage problem and to boost water/gas production in gas-water systems to the highest amount by changing contact angles and wettability. They illustrated that the gas production rate can be maximized using a favorable grade of contact angle. According to their mathematical model in terms of an even state of wettability, gas production will be maximized if the water contact angle is adjusted to 90° and 180° for low and high capillary-viscous ratios, respectively. And 90° was noted to be the optimum contact angle during piecewise wettability change in water-wet rocks regardless of capillary-viscous ratios. However, the model was theorized by assuming that the flows were incompressible, and it could not be applied in certain cases. This issue was tackled later when the above-mentioned research team extended their work by presenting a model that took the compressibility of gas into account, and the existence of an optimum wettability state was again deduced.<sup>[109,110]</sup>

Naik et al.<sup>[110]</sup> explained that as a result of wettability adjustment to gas-wet and increase in contact angles, wetting water will be declined or removed and productivity will increase, but gas and condensate will be moved to tighter pores, and this will reduce their production.<sup>[111]</sup> Thus, there needs to be an optimal contact angle in which the positive effect of wetting water reduction/removal eclipses the negative effect of gas displacement into less porous areas, and productivity will be maximized.

In further evaluations, Naik et al.<sup>[112]</sup> introduced the formation's wettability (water-wet or water-repellent), water cut, capillary-viscous ratio, and flow rate as factors that contribute to an optimum contact angle and eventually to the highest presumable productivity index (PI) in gas reservoirs. The importance of capillary forces, viscous forces, and flow rates was later elaborated by Reis and Carvalho<sup>[113]</sup> when they elucidated that even if the wettability is altered to a gas-wet state, gas

relative permeability may still reduce under the circumstance of low flow rates. Naik et al.<sup>[111]</sup> noticed that regardless of a formation's wettability, there is an opposite relationship between the amount of water cut and the optimum water contact angle. The same relationship was noted to exist between flow rate and optimal contact angle in hydrophobic formations, and the link between flow rate and optimal contact angle was found to be direct in hydrophilic formations.

Ajagbe et al.<sup>[114]</sup> referred to the possibility of an optimum wettability state existence by stating the idea that pores should be liquid-repellent enough for the condensate saturation to be reduced, and not gas-wet enough for the gas flow to be hindered. To prove this, they formed and ran 18 distinct CMG simulation cases by combining three different reservoir fluid properties (lean, intermediate, and rich), low and high permeability factors, and three wettability conditions (liquid-wet, intermediate gas-wet, and gas-wet). They concluded that high permeability reservoirs are not suitable candidates for wettability alteration unless the drainage area is on the high end. In the case of low permeability reservoirs, wettability alteration to intermediate gas-wetting condition is the best approach as it not only maximizes the production but also is economically beneficial compared to wettability change from liquid-wet to super-gas-wet state in terms of treatment expenses. Their study was in agreement with the research of Zoghbi et al.<sup>[115]</sup> who announced intermediate gas-wet as the most suitable wettability condition in terms of production maximization. This was later made evident by Baghnoaei et al.<sup>[116]</sup> who also researched treatment radius as a variable that impacts gas and condensate recovery along with wettability alteration.

The results of Naik et al.<sup>[111]</sup> and Ajagbe et al. were also confirmed by El Cheikh Ali et al.<sup>[117]</sup> By imbibition tests, flow tests, and radial simulations, El Cheikh Ali et al. approved the existence of an optimum wettability state on which the gas production will be maximized and beyond which it will be declined. In other words, with regard to changing wettability to gas-wet conditions, they disproved the theory that the higher the liquid contact angle, the higher the production yield. They added that the optimum wettability is reached when the gas end-point relative permeability increases and the average oil saturation drawn from imbibition tests in wettability-altered cores becomes higher than authentic ones. They announced intermediate gas-wetting as the most productive state to be targeted in low permeability reservoirs. They also discouraged wettability alteration treatments for high permeability reservoirs due to the agglomeration of heavy condensate components in the reservoir. It must be mentioned that Ajagbe et al. and El Cheikh Ali et al. utilized chemical substances as treatments and nanoparticles were not used.

#### **4. Analytical discussion**

SiO<sub>2</sub> NPs are the most commonly used type of NPs for gas-wetting alteration of condensate reservoirs because of their low price, abundance, endurance in high-pressure high-temperature (HPHT) conditions, and low toxicity. Furthermore, if properly functionalized, these NPs are effective on all kinds of reservoir rocks in terms of gas-wetting alteration. Surfapore M is also



proven to be applicable in both carbonate and sandstone reservoirs. Several studies reported in this article already emphasized on the surpassing ability of NPs to alter the wettability of reservoirs to gas-wet compared to other chemicals. However, it must be noted that the superiority of NPs to chemicals is not absolute and is utterly case and material dependent. A vivid example of this was demonstrated by Sayed et al.<sup>[6,54]</sup> when they developed an F-SiO<sub>2</sub> nanofluid that did not surge the relative permeability of gas and condensate as much as a fluorinated polymeric surfactant used by Al-Yami et al. Despite this, treating samples with F-SiO<sub>2</sub> NPs was a more durable approach and thus possibly a more cost-effective approach than treating them with the aforementioned fluorinated polymeric surfactant. In general, we cannot declare a certain nanoparticle as the best one to be used in gas-wetting alteration by only comparing the research results of various scholars mentioned in this article. This is due to the large number of variables existing in a comparison. In other words, the results of different research teams can not be compared because different rocks and concentrations of nanofluids were employed in each experiment. Moreover, the procedure and conditions of each experiment conducted to assess the extent of wettability alteration are different. These differences were already mentioned for the contact angle test. Similar differences exist in coreflooding and imbibition tests conducted by different researchers.

To concisely assess and confirm the effectiveness of wettability alteration on the alleviation of liquid blockage in experiments, it is essential to simulate condensation and include liquids that are representative of condensate in contact angle and flow tests (coreflooding, visualizations of flooding in micromodels, sand pack flooding). Not using condensate in coreflooding tests of Tang and Firoozabadi<sup>29</sup> and Fahes and Firoozabadi's<sup>[118]</sup> research was criticized by other researchers.<sup>[119,120]</sup> Using actual condensate in the mentioned experiments can demonstrate the capabilities of NPs in the most accurate way and is notable as an advantage of several studies already reported in this paper.<sup>[53,59,62,79-81,91,92]</sup> Neither condensate nor its representatives were employed by Wang et al.<sup>[89,90]</sup> but this can not be criticized because their research aimed to eliminate only water blockage and not other liquids in general. However, since they concluded wettability improvement to gas-wet state, contact and flow tests utilizing condensate can be performed to more directly examine the performance of CNDAD1# nanofluid in condensate reservoirs in future research. Undoubtedly, conducting adequate tests to check the effectiveness of nanofluids can not be neglected; Shayan and Esmailnezhad illustrated the performance of nanofluids via only contact angle tests and no flow tests were conducted. Neither flow nor contact angle tests were carried out by Ganie et al. However, these research teams introduced novel nanofluids to the literature and we encourage a more direct investigation of the capability of NPs promoted by these scholars by imbibition and flow tests.

With respect to the wettability state in the pore scale, water wettability causes water molecules to adhere to pores and block them, and thus production declines. The same blockage and subsequent decline in production occur for oil wettability by oil molecules. In the state of absolute gas wettability, gas molecules adhere to pores which facilitates water and oil production. This slightly hinders gas production in tight pores. Nevertheless, the negative impact of gas wettability on gas

and condensate production is negligible compared to the adverse impact of water and oil wettability. Because of this, as covered in the previous section, intermediate-gas wettability has been proven to be the condition that maximizes gas and condensate production. Despite this, obtaining gas or super gas-wet surfaces was the target of the majority of research already mentioned in this paper. This approach is unexceptionable. If gas/super gas-wetting alteration is considered a transient but stable treatment whose effect will gradually decrease so that the super gas-wet area around the wellbore descends to an intermediate gas wet one. Under this circumstance, if only the productivity (and not economical) aspect of the reservoir is of concern, targeting a super gas wet state can be more beneficial than an intermediate gas wet state since it lasts longer and production is higher compared to water and oil wetting states. Esmailzadeh et al.<sup>[121]</sup> developed potent NP coatings on carbonate rocks that would completely retain the super gas-wet properties in temperatures up to 180 °C; these properties would only begin to diminish after nearly 16 cycles of erosion with 240-grit sandpaper. Their results can be remarked as an apt example of a highly stable, but transient super gas-wet state. Sayed et al.<sup>[65]</sup> revealed that some modified NPs form more durable gas-wet surfaces than others. These findings along with laboratory experiments conducted by other research teams on the durability of the gas-wet layer on rocks' surfaces indicate that it is possible for a super gas-wet coating to descend to an intermediate gas-wet one or completely wear off. The degree of the wear off of the gas-wet coating formed by NPs depends on NPs, components that modify NPs, reservoir conditions (temperature, salinity), and production conditions such as flow rate. Hence, this degree can be low enough for the super gas wet state to be considered stable and permanent. In this case, aiming a super gas wettability in the treatment is not economical. However, due to harsh reservoir conditions in some cases, the achieved wetting state can be considered temporary. Thus, forming a super gas-wet state may lead to higher production amounts.

Most importantly, we pose two novel notions concerning the gas-wetting alteration of condensate reservoirs via NPs. Compared to liquids, employing foam as the carrier of wettability enhancement chemicals is advantageous. As a result of injecting chemicals in foams, less liquid will be used. Apart from economical aspects, this reduces clay swelling and the chance of liquid bridges will be decreased. Hence, there is a higher possibility for permeability to remain intact. The use of foam will also channel injected gas and chemicals into tighter pores which results in better sweep efficiency. NPs have been employed in foams as stabilizers. Foams formed with surfactants have also been utilized to change reservoir wettability. However, the literature lacks an assessment of the wettability alteration of condensate reservoirs via a foam formulation in which modified NPs are included as the main wettability changers of the surface and not as foam stabilizers. Such an approach may be also described as a combination of gas injection and wettability alteration to produce built-up condensate. Another theory is to use NPs to alter the wettability and hinder sand production altogether. As it was noted, SiO<sub>2</sub> NPs have been applied to prevent sand production. They have also been vastly used in wettability alteration cases. We believe that it is possible to apply silica (or other types of) nanoparticles to change wettability and prevent sand production in oil and condensate reservoirs at the same time. This seems possible due to the similar nature of

required materials in both wettability alteration and sand production prevention which are materials that should not reduce permeability. However, since this has never been tried before, this theory must be assessed by physical and numerical experiments.

## 5. Conclusion

The most noteworthy research papers about the wettability alteration of retrograde gas condensate reservoirs using nanoparticles were categorized and highlighted, and their results were compared. In terms of wettability alteration, nanoparticles are generally more effective than fluoropolymers and other chemicals. This is demonstrated by nanoparticles' surpassing thermal resistivity and fewer volumes of nanosolution required for treatments. Until a certain temperature, the nanoparticles' capability to form gas-wet surfaces improves with the increase in temperature. While initial wettability condition, oil saturation, and salinity can negatively affect the performance of nanoparticles, the final results always confirm a successful treatment. Different conditions of experiments conducted in the investigations reported in this article act as limitations that do not let us directly compare and rank different nanoparticles and approaches in terms of gas-wetting alteration ability. Hence, more research is required to compare the effectiveness of different nanoparticles in identical conditions in the future. In addition to promoting current usages on larger scales, this review provided an outlook of possible future applications of nanoparticles, and the following conclusions were highlighted:

- While the effectiveness of all the tested nanoparticles has been proven, silica nanoparticles are the most commonly used type in changing the wettability of gas condensate reservoirs from liquid wet to gas wet because of their effectiveness, competitive price, and resistance against high temperature and pressure.
- The optimum wettability condition that should be targeted in the treatment is case-dependent. If the reservoir environment is harsh, forming a super gas-wet state may be the best target and otherwise reaching an intermediate gas-wet state maximizes production. In terms of super gas wettability, economical aspects must be considered as well.
- Nanoparticles can be injected into reservoirs using foam-based carriers. In this way, they can act as both foam stabilizers and wettability alteration agents and less formation damage and more surface treatment occur due to the nature of the foam.
- Nanoparticles can be applied to decrease fines migration and sand production and abolish liquid blockage altogether.

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## Conflict of interest

The authors have no competing interests to declare that are relevant to the content of this article.

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