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Review

Fundamental Mechanisms and Factors Associated with Nanoparticle-Assisted Enhanced Oil Recovery

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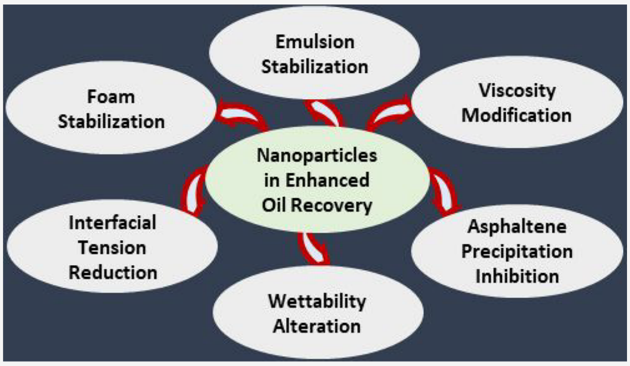
ABSTRACT: Nanoparticles have been used in conventional enhanced oil recovery to improve the oil recovery efficiency. In this review, we analyzed the roles of nanoparticles in improving enhanced oil recovery. We focused on elucidating the fundamental mechanisms that regulate nanoparticle-induced enhanced oil recovery, including surface wettability alteration, interfacial tension reduction, mobility ratio increase, emulsion stabilization, and foam stabilization. We also discussed factors that control the success of nanoparticle-induced enhanced recovery. Finally, we outlined the current challenges and future research directions with respect to the applications of nanoparticles in enhanced oil recovery.



1. INTRODUCTION

The U.S. Energy Information Administration (EIA) has predicted that worldwide energy consumption will increase by 50% before 2030.[1] The consumption of hydrocarbon energy (i.e., petroleum oil and gas), one of the main fossil fuel energy sources, has been increasing steadily during the past few years.[2,3] There are three phases of oil recovery in practice in the oil industry, including primary, secondary, and tertiary, or enhanced oil recover (EOR).[4] After the primary and secondary recovery operations, around 60%−70% of the original oil still remains in place.[5] As opposed to mainly relying on the displacement energy in the primary and secondary recovery processes,[6] various EOR methods were designed to improve interactions between the rock/oil system and the injected fluids or thermal energy. In recent years, nanoparticles have been used to improve conventional EOR techniques.[7,8] Nanoparticles are defined as particles with diameters equal to or less than 100 nm. The unique properties of nanoparticles, such as high surface-to-volume ratio, modifiable surface properties, magnetic responsiveness, pH responsiveness, and environmental friendliness, make them advantageous for EOR applications. Suspensions of nanoparticles (i.e., nanofluids) are produced in a base water, oil, gas, or surfactant solution.[9,10] Recent laboratory experiments have suggested that the injection of nanofluids has the potential to recover about 50% of the oil that remains in the reservoir after the primary, secondary, and even conventional EOR processes.[11−14]

Various types of mechanisms are involved in nanoparticle-assisted EOR. Nanoparticles used in surfactant flooding help reduce the interfacial tension (IFT) and better withstand harsh reservoir conditions.[15,16] Nanofluids change the wettability of rock surfaces by creating the disjoining pressure and adsorption onto the reservoir matrix.[10,17] Nanoparticles have been widely used to modify the viscosity of oil, improve the mobility ratio, and address issues such as viscous fingering and early breakthrough.[18−20] Pickering emulsions (i.e., nanoparticle-stabilized emulsions) provide better emulsion stability and emulsifying capacity even under harsh reservoir conditions.[21−23] Foams stabilized by nanoparticles show stability against interbubble gas diffusion, liquid drainage, and coalescence.[24−26] Laboratory-scale success of nanoparticles in foam and emulsion stabilization,[27,28] wettability alteration and IFT reduction,[29,30] and mobility control[31] has made them promising for field-scale applications. The successful application of nanoparticles in EOR is dependent on an advanced understanding of the mechanisms that regulate the fundamental processes in nanoparticle-induced EOR. This review provides a comprehensive summary on fundamental mechanisms associated with the nanoparticle-assisted EOR.

Over the past years, some previous reviews presented the successes and challenges of the applications of nanoparticles and nanotechnology in the petroleum industry.[8,12,16,32−41] Many of these reviews have discussed mechanisms associated

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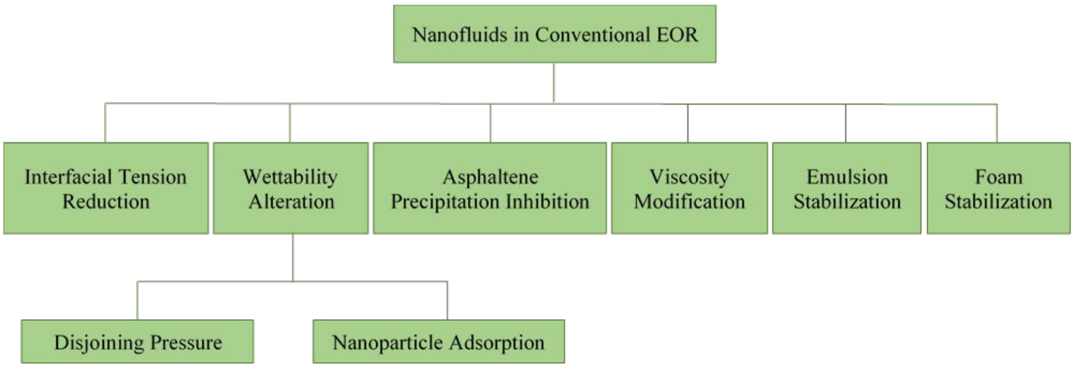

Figure 1. Nanofluid contributions to conventional EOR.

with nanoparticle-assisted EOR.[12,36,37,41−45] However, discussion on how nanoparticles regulate a mechanism is typically not a central thread. Combinatorial applications of nanoparticle−surfactant[43,45,46] and low salinity water−nanoparticle[43] were also reviewed. A few reviews described general mechanisms in the nanoparticle-induced EOR, but were more focused on comparing the efficiencies of various types of nanoparticles in EOR applications.[12,41,42,44] Sometimes the reviews were focused on only one or two specific mechanisms.[12,32,37,38,41,42,44,45] A few other reviews[16,32,35] focused on the applications of nanotechnology in various stages of oil production, including exploration, reservoir characterization, drilling, cementing, scaling control, and various types of EOR. Some reviews emphasized the use of various types of nanoparticles for formulating nanofluids,[8,12,42,47] properties and stabilities of nanofluids,[12,40,42,46] and methods used in nanofluids' property measurements.[40] A few focused on one or two types of nanoparticles, such as the application of bare nanoparticles (i.e., nanoparticles without surfactant or polymer),[34,41] polymer-coated nanoparticles,[33,47] and polymeric nanofluid suspensions[47] in EOR. A few others discussed the application of nanoparticles in a particular EOR mechanism, such as emulsion stabilization[38,48] and foam stabilization.[49]

However, a review focused solely on the state-of-the-art understanding of the fundamental mechanisms and factors associated with nanoparticle-induced EOR processes is missing. So far, significant research advancements have been made with respect to the applications of nanoparticles in EOR. Therefore, a comprehensive review on this topic is necessary and crucial. In this review, the roles of nanoparticles in wettability alteration, IFT reduction, mobility control, emulsion stabilization, and foam stabilization are discussed and summarized. Factors that govern a particular mechanism are discussed, and suggestions are made to facilitate nanoparticle application for that specific mechanism. A relative comparison of nanoparticle efficiencies in EOR was widely discussed in other published reviews;[12,41,42,44] therefore, that discussion is not a part of this review. Figure 1 illustrates the overall structure of this paper and the contributions of the nanofluids in conventional EOR processes.

2. NANOPARTICLE INDUCED OIL−WATER INTERFACIAL TENSION REDUCTION

IFT refers to the attractive force between the interfacial molecules of two immiscible liquids, which is usually expressed in the unit of dyn/cm or mN/m.[50] IFT is an important parameter in determining the distribution and movement of fluids in porous media.[45] A lower IFT leads to a higher ratio of viscous force to capillary force (i.e., the dimensionless capillary number, Ca), which can increase the efficiency of EOR.[45] Surfactant flooding has a long application history of reducing the IFT. However, it is reported in the literature that some reservoir conditions, including high temperature, salinity, and reservoir formation, limit the use of surfactant flooding in EOR.[51,52] For instance, anionic surfactants can readily adsorb onto carbonate reservoirs. Cationic surfactants could be an alternative to anionic surfactants due to their lower adsorption tendencies in the carbonate reservoir, but their high cost limit the application. Therefore, the application of surfactant flooding is not common in a carbonate reservoir.[7] However, the use of nanoparticles alone or in conjunction with surfactants proved to be very efficient in a carbonate reservoir.[53] Bayat and Junin showed a reduction in IFT and an improvement of 52.6%, 50.9%, and 48.7% in oil recovery by using Al_2O_3, TiO_2, and SiO_2 metal nanoparticles, respectively, in porous limestone media.[53] Although surfactants performed poorly in a highly saline environment due to the high precipitation rate,[7] Sulaiman et al.[54] showed silica nanoparticles could reduce the IFT and improve the oil recovery to 64.5% in highly saline porous media. Additionally, while high temperatures limit the use of surfactants due to high precipitation rates,[7] Esfandyari Bayat et al.[55] showed Al_2O_3, SiO_2, and TiO_2 nanoparticles could effectively reduce the IFT as temperature increases.

2.1. Mechanisms of Nanoparticle-Induced Oil−Water Interfacial Tension Reduction.

When nanoparticles are used with surfactants for IFT reduction, the kinetics and reduction rate depend on many factors, including nanoparticle concentration and wetting ability, surfactant charge, and concentration. Mechanisms of IFT reduction are different for hydrophilic and hydrophobic nanoparticles.

When hydrophilic nanoparticles are used with a very low concentration surfactant of similar charge, they get wetted by water and tend to reside in the bulk solution instead of accumulating at the interface; hence, they have a minimal effect on IFT reduction.[56,57] Zargartalebi et al.[58] used negatively charged hydrophilic silica nanoparticles in a solution with a very low concentration of anionic surfactant and found no IFT reduction. When the surfactant concentration approaches the critical micelle concentration (CMC), IFT can be reduced. The mechanisms controlling the IFT reduction depend on the charges of nanoparticles and surfactants. If nanoparticles and surfactants are similarly charged, and the concentration of the surfactant is not higher than CMC, and the electrostatic repulsion force will make the nanoparticles diffuse toward the oil−water interface and reduce the IFT.[59−62] However, if the surfactant concentration is higher than the CMC, the surfactant will completely occupy the interface, and the repulsion force will cause the particle to be in suspension.[58,60] Ma et al. observed an enhanced IFT reduction due to the synergistic effect of the negatively charged silica nanoparticle and anionic sodium dodecyl sulfate (SDS) surfactant and attributed the effects to the increase in the repulsive interaction between silica nanoparticles and SDS.[59] When nanoparticles and surfactants are oppositely charged, the hydrophilic heads of surfactants get adsorbed onto the surfaces of nanoparticles, leaving hydrophobic tails of surfactants floating in the solution.[63] The floating hydrophobic tails of surfactants pull nanoparticles toward the interface, which results in IFT reduction.[56] Ravera et al.[56] investigated the synergistic effect of a cationic CTAB surfactant and negatively charged silica nanoparticles on IFT and attributed the enhanced IFT reduction to the increased surfactant coverage on nanoparticles with an increase in surfactant concentration. The nanoparticles adsorption at the interface decreases the internal energy (U) of the system. The movement of larger particles toward the interface replaces the smaller electrolyte molecules at the interface, which leaves empty space within the system, resulting in an increased Brownian motion of electrolyte molecules and therefore increased system entropy (S).[15] As a result of the rise of entropy and a decrease in the internal energy, the total free energy, defined as $F = U - TS$ decreases, where T is the temperature.[15] The thermodynamic definition of IFT is described in eq 1[15]

$$\mathrm{IFT} \equiv \frac{\partial F}{\partial A} \tag{1}$$

where A is the surface area. According to eq 1, a decrease of F will lead to a decrease in IFT. This is true when the surfactant concentration is lower than CMC.[64] Interestingly, when the surfactant concentration is higher than CMC, IFT will continue to increase until it reaches a plateau after a particular concentration (Figure 2). This phenomenon was nicely explained by Dong and Johnson[15] using a capillary force concept (Figure 3). A further increase in the surfactant concentration will push the existing nanoparticles to partially float (Figure 3a) due to the floating hydrophobic tails of surfactants attached to nanoparticles or to immerse (Figure 3b) at the interface due to electrostatic repulsion if nanoparticles and surfactants are similarly charged. The capillary force is significant for immersed particles, even for very small-sized particles.[15] When the nanoparticle concentration at the interface reaches high enough to form multipole attraction (Figure 3c), the capillary force reaches very high to resist the deformation, which can result in the increase of IFT (Figure 2).[15] However, when the nanoparticle concentration at the interface reaches saturation, the capillary force will not continue to increase; hence, the IFT will not increase further (Figure 2).[15]

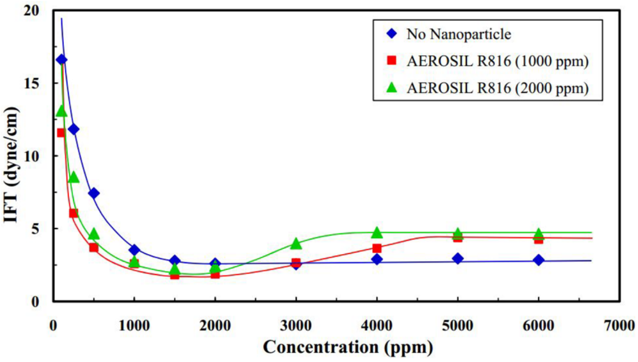

Figure 2. Results show the change in IFT when only SDS is present versus the synergistic effect of SDS and hydrophobic silica nanoparticles, which is commercially known as AEROSIL R816. Reproduced with permission from ref 58. Copyright 2014, Elsevier.

Partially hydrophobic nanoparticles, in conjunction with surfactants, can also contribute to IFT reduction,[58] although the mechanism is different than hydrophilic nanoparticles and surfactants. Zargartalebi et al.[65] observed a similar trend in IFT reduction for partially hydrophobic and hydrophilic silica nanoparticles. However, at a low surfactant concentration, partially hydrophobic nanoparticles show a noticeable impact on the IFT reduction, whereas hydrophilic nanoparticles do not.[58,65] This is because partially hydrophobic nanoparticles have a high affinity to the oil−water interface due to their amphoteric natures, which is not dependent on surfactant concentration. On the other hand, hydrophilic nanoparticles have the tendency to reside in the bulk phase, and the surfactant concentration is not high enough to bring or push nanoparticles to the interface at a low surfactant concentration. Similar to hydrophilic nanoparticles and surfactant mixtures, IFT can be reduced when the surfactant concentration increases up to CMC when mixing with partially hydrophobic nanoparticles. This phenomenon happens due to the formation of a cluster between the partially hydrophobic nanoparticle and the hydrophobic tail of the surfactant. A further increase of surfactant concentration beyond CMC results in the increase of IFT, due to the capillary force concept explained in Figure 3.[15] However, there is a lack of research on the synergistic application of hydrophobic nanoparticles and surfactants in IFT reduction.

Nanoparticles are predominantly used in conjunction with surfactants for the oil−water IFT reduction in EOR applications. Recently, nanoparticles have been used solely in the oil−water IFT modification. Studies reported the decrease,[66−69] increase,[70] and even no change[56,71] in the IFT owing to the use of nanoparticles.[72,73] It has been proved from experimental results and thermodynamic models that the Brownian motion can redistribute the nanoparticles at the liquid phase or interface where the particles' interaction is most favorable and the total free energy is lowest.[73−76] The decrease in the IFT happens due to the adsorption of the nanoparticle at the interface. Sometimes, even after the adsorption of the nanoparticle at the interface, no change occurs in the IFT because the particles are not surface active.[73,77] The increase in the IFT in the nanoparticle's presence is probably due to the interaction of the liquids; however, the mechanistic reason behind this is not clear yet.[70] Particle adsorption energy (E_d), the energy required to remove particles from the interface, can be calculated using the equation[73,74,78] $E_d = \pi R^2 \gamma_{ow} (1 - |\cos\theta|)^2$, where R is the nanoparticle radius, γ_{ow} the interfacial tension at the oil−water interface, and θ the contact angle. A contact angle of 90° is most favorable for the particle's adsorption at the interface, and an increase or decrease from 90° will facilitate the movement of the particles into the bulk phase. The particle's adsorption or desorption energy at the interface also depends on the particle's surface charge density, which can be influenced by electrolyte concentration or

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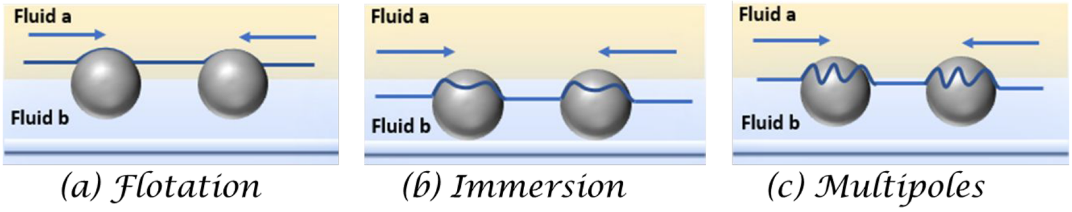

Figure 3. Schematic of the capillary force concept. (a) Particles are freely floating at the interface, (b) particles are partially confined in the liquid, and (c) the contact line of the particle surface is irregular, resulting in a higher particle concentration than (a) and (b). Schematic was drawn with the idea from ref 15.

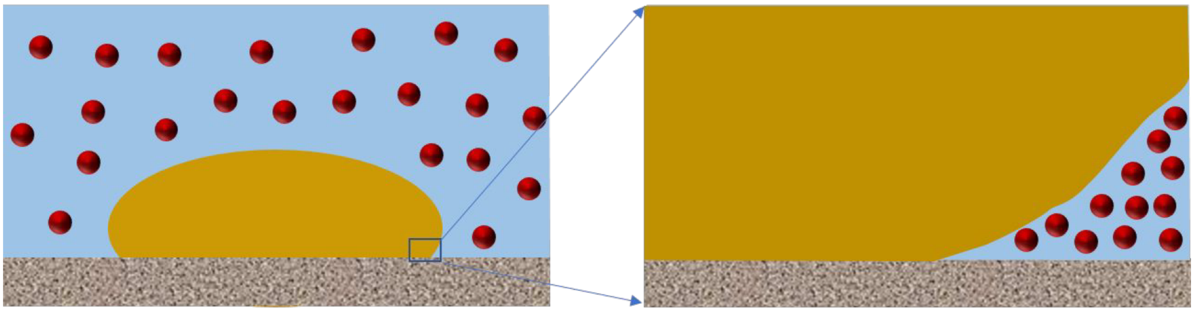

Figure 4. Wedge-film forming in a three-phase confinement. Schematic was drawn with the idea from ref 95.

solution pH.[79,80] An increase in the surface charge density will increase the electrostatic repulsion force, facilitating the particle's desorption from the interface.

2.2. Factors That Affect Oil–Water IFT Reduction. Nanoparticle surface wettability (i.e., hydrophobicity and hydrophilicity) is the most critical factor that influences the arrangement of nanoparticles at the oil–water interface.[58,81] The electrolyte concentration influences the wettabilities of the nanoparticles[82,83] and stabilities of the surfactants.[84] The type of rock in the reservoir impacts the mobility of the surfactant and the selection of surfactants for IFT reduction.[85] In addition, reservoir temperature also affects the surfactant or nanoparticle performance in IFT reduction.[86] If hydrophilic nanoparticles are used in conjunction with surfactants, the surfactant concentration plays an important role in determining nanoparticle efficiency in IFT reduction.[58,59] However, the hydrophobic nanoparticle can contribute to the IFT reduction regardless of the surfactant concentration.[65] In both cases (hydrophilic and hydrophobic nanoparticles), an increase in the surfactant concentration up to the CMC positively influences the IFT reduction.[59]

3. WETTABILITY ALTERATION

Reservoir formations can be water-wet, oil-wet, or intermediate-wet.[87] Waterflooding is a well-known method to increase oil recovery efficiency following primary production based on the spontaneous imbibition of water.[88] Spontaneous imbibition occurs at high capillary pressure (Pc), which is defined as the pressure difference between the nonwetting fluid and the wetting fluid

$$\mathrm{Pc} = \mathrm{Pn} - \mathrm{Pw} \tag{2}$$

where Pn is the nonwetting phase pressure, and Pw is the wetting phase pressure.[89] Capillary pressure is negative in oil-wet reservoirs, so water imbibition does not occur spontaneously.[82,90] Hence, it is crucial to alter oil-wet surfaces into water-wet or intermediate-wet surfaces, so that imbibition of water can occur. Altering the wettability to water-wet has proven to be very efficient in increasing the spontaneous imbibition of water, which ultimately displaces the oil in the reservoir matrix.[91]

Nanomaterials have been used to enhance the efficiency of wettability alteration. Here, we reviewed the mechanisms of two types of technologies that use nanoparticles to enhance the efficiency of wettability alteration, i.e., flooding nanofluids and flooding surfactant and nanoparticle mixtures.

3.1. Wettability Alteration by Nanofluid Flooding. Nanofluids have several advantages over traditional chemical methods on wettability alteration, including high efficiency, and most instances low cost, and environmental friendliness.[92,93] The mechanisms that regulate wettability alteration and nanofluid spreading on the solid surface are different from those for Newtonian fluids. For nanofluids, an increase in the nanoparticle concentration will result in an increase in the viscosities of fluids, which should decrease the fluid spreading on the surface. However, previous experimental studies showed that an increase in the nanoparticle concentration up to a specific concentration resulted in increased spreading.[17,94] This indicates that all the well-established theories of Newtonian fluid spreading on the solid surface are not applicable in the case of nanofluids.[95] The mechanisms of nanofluid spreading and wettability alteration have been investigated by researchers for a long time, but they have not been fully understood yet. So far, two possible mechanisms are widely accepted: (1) structural disjoining pressure[10] and (2) nanoparticle adsorption at the solid–liquid interface.[17] Wasan and Nikolov[10] proved that the structural disjoining pressure resulting from the formation of a wedge structure (Figure 4) at the three-phase contact region was the primary mechanism behind the wettability alteration by nanofluids (first mechanism).[93] Nanoparticle adsorption in the reservoir system was proven to be another mechanism in wettability alteration (second mechanism).[17,19]

3.1.1. Structural Disjoining Pressure. Wasan and Nikolov[10] first demonstrated that nanofluids form a wedge-like film at the confined three-phase contact region (i.e., oil drop on a solid surface with nanofluids) (Figure 4). Nanoparticles confined between the oil drop and solid surface are known to self-order into one, two, or three layers (Figure 5),[96] which increases the entropy of the confined film and promotes additional nanoparticle movement. The increased nanoparticle movement raises pressure at the three-phase confinement, which is defined as the disjoining pressure.[10,95] Disjoining pressure includes several components, including van der Waals force, electrostatic force, solvation forces, and structural disjoining pressure.[95,97] Several disjoining pressure components (for example, van der Waals, electrostatic, and solvation forces) are only prevalent at short spatial scales (i.e., scales shorter than the effective diameter of the nanoparticle).[95] The structural disjoining pressure dominates at larger scales (i.e., scales larger than the effective diameter of a nanoparticle).[98] The structural disjoining pressure is considered the most important factor that controls the spread of nanofluids and alters the wettability.[17,97] Some studies[10,97] found that a decrease in the film thickness can increase the spreading coefficient, a parameter estimated based on the structural disjoining pressure. As the number of layers in the wedge film decreases, the disjoining pressure increases, or the disjoining pressure is higher in the region of one layer than in the region of two layers in the wedge film. Thereby, the disjoining pressure is higher at the wedge tip (three-phase contact line), which facilitates the spreading of the nanofluid and alters the wettability from the oil-wet to the water-wet. Sefiane et al. observed an increased spreading of alumina nanoparticles on a hydrophobic surface when the alumina nanoparticle concentrations were up to 1 wt %.[17] However, a further increase in the concentration of alumina nanoparticles resulted in a decrease in spreading, or no change in spreading happened. The authors explained the phenomenon based on observed changes in contact angle with the presence of smaller-sized nanoparticles.[94] Briefly, with the presence of smaller-sized nanoparticles, a large number of particles could squeeze into the wedge film due to their smaller size, which caused a greater change in the contact angle. On the other hand, an increase in the concentration could have stimulated higher particle—particle interactions, thereby resulting in the formation of a large agglomeration. The large size of the agglomeration limited the number of particles that could reside at the interface and hence decreased the spreading and wettability alteration capacity of the nanofluids. The relation between the nanoparticles' sizes, film energies, and spreading is proved from both experimental and modeling studies.[98−101] Kondiparty et al.[99] showed that at the same concentration (i.e., 25 vol %), the number of particles in the film was 4.3 times higher for the 18.5 nm than 30 nm particles. A higher number of particles in the film results in a higher structural disjoining pressure and higher nanofluid spreading.[99−101]

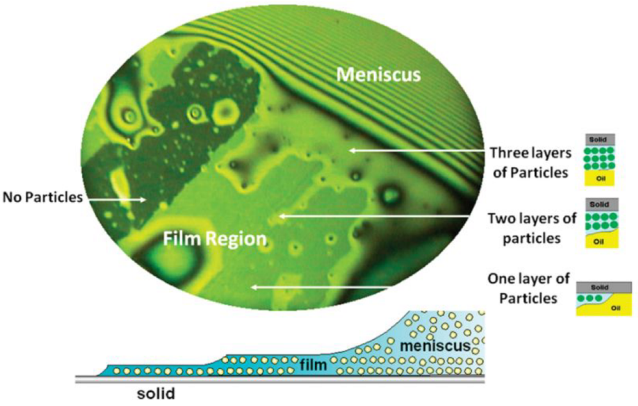

Figure 5. Photomicrograph depicting the nanoparticle layering of 10 vol % aqueous silica suspension (19 nm nominal particle diameter) on a solid surface (film size ≃ 868 μm). Reproduced with permission from ref 96. Copyright 2010, American Chemical Society.

3.1.2. Nanoparticle Adsorption at Solid−liquid Interface. Nanofluid flooding can lead to nanoparticle adsorption at the solid−liquid interface, which is another possible reason for the wettability alteration (Figure 6). Sefiane et al.[17] hypothesized that the adsorption of nanoparticles on the surface provides a lubricating effect due to the sphericity of the nanoparticle, which facilitates the spreading of polydisperse alumina nanoparticles on surfaces. Due to the lubricating effect and smaller friction, liquids tend to slip as if on a superhydrophobic surface.[102] Maghzi et al.[19] used a micromodel to investigate the mechanisms of wettability alteration by hydrophilic silica nanoparticles. They found that the adsorption of nanoparticles on surfaces increased the surface free energy and modified the surface from oil-wet to water-wet. An increase in the nanoparticle concentration increased the oil recovery up to the concentration sufficient to saturate the available adsorption sites. However, an increase in the concentration from the saturation concentration would result in a decrease in the oil recovery due to the blockage of pores and throats.[19] Besides the nanoparticle concentration, the nanoparticle adsorption is also influenced by the change in the salinity and exposure time. A higher salinity or exposure time resulted in high adsorption and wettability alteration.[103]

3.2. Wettability Alteration by Flooding Surfactant and Nanoparticle Mixtures. Surfactant flooding can change

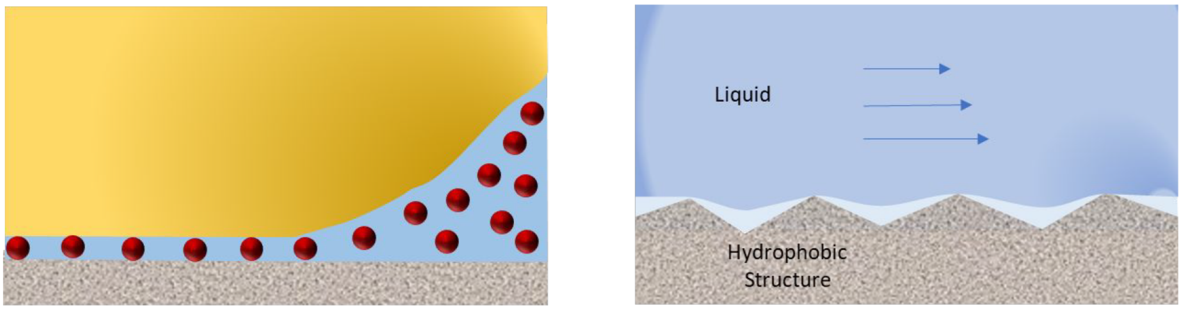

Figure 6. Wettability alteration due to nanoparticle adsorption (left) and liquid sliding on the nanostructured surface (right). Schematic was drawn with the ideas from refs 17 and 102.

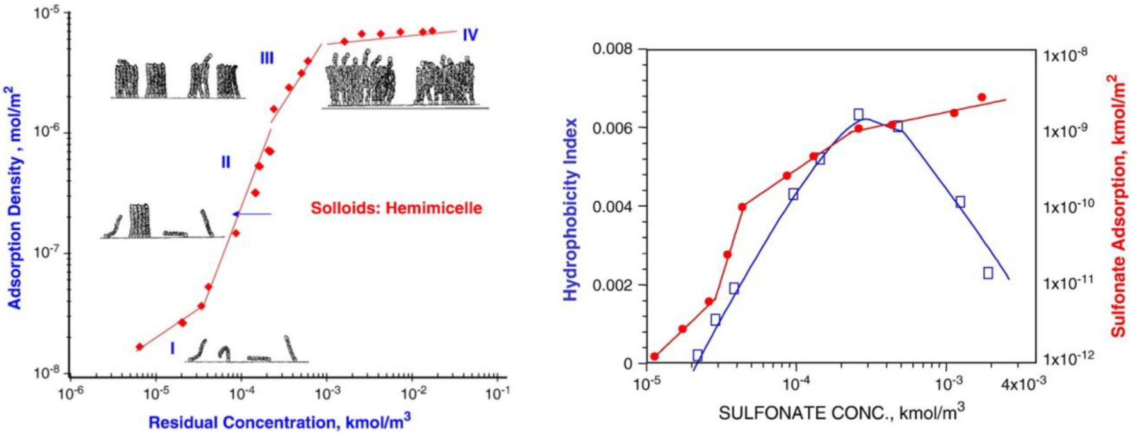

Figure 7. Schematic representation of the hypothetical general trend of the surfactant's adsorption isotherm with four different regions (left) and n-dodecyl benzenesulfonate adsorption and its effect on the hydrophobicity (right). Reproduced with permission from ref 108. Copyright 2006, Elsevier.

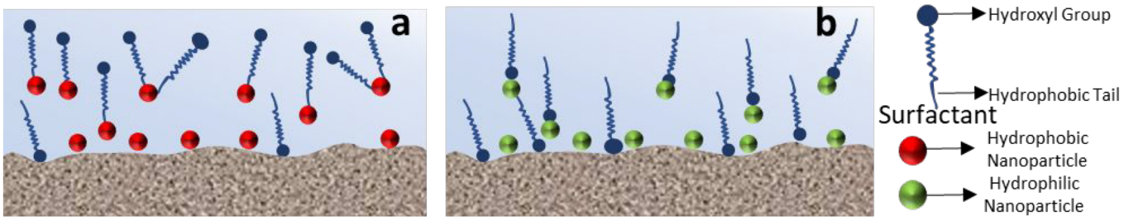

Figure 8. Schematic representation of the nanoparticle-induced surfactant adsorption for (a) hydrophobic nanoparticles and (b) hydrophilic nanoparticles. Schematic was drawn with the idea from ref 112.

the wettability and increase spontaneous imbibition, enhancing the oil recovery factor.[88,104] Although surfactant flooding increases the efficiency of EOR, surfactants tend to adsorb to rock surfaces. The loss of surfactant due to sorption can decrease the surfactant concentration, making the process less efficient over time for EOR application.[105] Mixing nanoparticles with surfactants could decrease the adsorption of surfactants on rock surfaces.[106,107]

Adsorption of surfactants on solid surfaces is controlled by several processes, including electrostatic attraction or repulsion, hydrophobic interaction, hydrogen bonding, ion exchange, chemisorption, and chain−chain interaction.[108] Somasundaran and Fuerstenau[109] proposed a four-region surfactant adsorption mechanism, as illustrated in Figure 7. Initially, in region I, when the rock surfaces were free from any surfactant, adsorption was solely due to the electrostatic interaction between the charged headgroups of surfactants and the mineral charged surfaces.[108] In region II, adsorption sharply increased due to two simultaneous processes: (1) electrostatic interactions between the free charged mineral surface (i.e., mineral sites which did not attach any surfactants in the region I) and the charged headgroup of the surfactant and (2) hydrophobic interactions between the adsorbed surfactant tails and tails of the free surfactant in the solution.[110] The hydrophobic interactions can take place far below the CMC, and the aggregates formed due to hydrophobic interactions are called solloids (hemimicelles).[108,111] In region III, the adsorption rate gradually decreased, which could be attributed to the charge neutralization of the rock surface due to the adsorption of surfactants in regions I and II. In addition, the adsorbed surfactants created charges on rock surfaces, which prevented further adsorption of surfactants with similar charges.[108,110] Although the adsorption rate decreased, some adsorption still occurred in region III due to surfactant chain−chain hydrophobic interactions. In region IV, the adsorption reached a plateau because the surface was already completely covered, and the surfactants in the bulk phase formed micelles.[110]

The surfactant adsorption continuously modified the wettability of the rock surface, as illustrated in the right panel of Figure 7. With the increase of surfactant concentration, rock surfaces that were initially hydrophilic gradually changed to hydrophobic. The hydrophobicity of the surface reached a peak at the end of region II. In region III, the hydrophobicity level decreased due to the exposed hydrophilic headgroup of the adsorbed surfactant.[108] For the same reason, the decreasing trend in hydrophobicity extended to regions III and IV.

The presence of nanoparticles in surfactants can change CMC and surfactant adsorption behaviors. Ahmadi and Shadizadeh[112] investigated the effects of silica nanoparticles on the adsorption of surfactants on sandstone rock surfaces (Figure 8). They found that both hydrophilic and hydrophobic nanoparticles can reduce the adsorption of surfactants on rock surfaces. Hydrophilic nanoparticles reduce surfactant adsorption by means of three mechanisms. First, hydrogen bonds can form between hydroxyl groups of the surfactants and hydroxyl groups of the hydrophilic nanoparticles, which helps the surfactant remain in the suspension.[112] Second, silica nanoparticles can adsorb onto rock surfaces due to the electrostatic attraction force between the mineral charge and nanoparticles charge and form a nanoparticle layer, inhibiting further adsorption in the interface.[89,107,113] Third, the collision between the attached surfactants and flowing nanoparticle

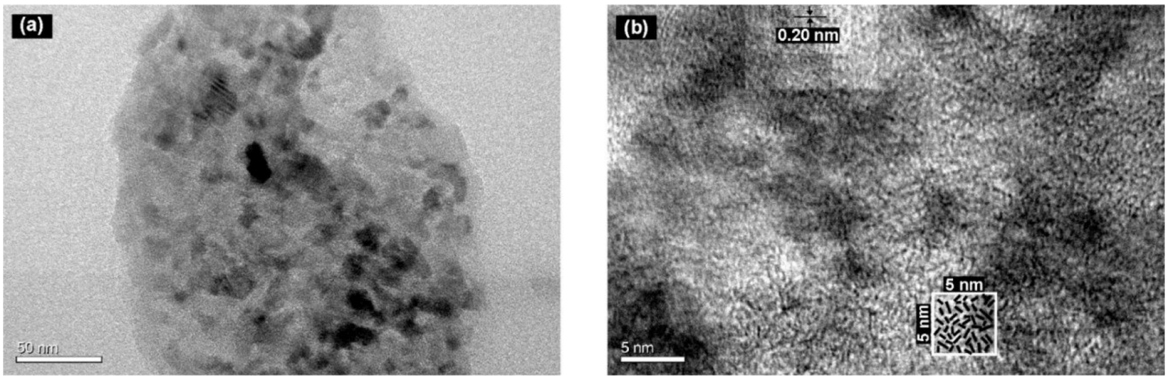

Figure 9. HRTEM images showing asphaltene adsorbed in MgO nanoparticle: (a) 50 nm and (b) 5 nm scale bar. Reproduced with permission from ref 123. Copyright 2013, American Chemical Society.

suspension can result in the detachment of the attached surfactants.[107] In the case of hydrophobic nanoparticles, the hydrophobic tails of surfactants are attracted by hydrophobic nanoparticles due to hydrophobic interactions, which reduce surfactant adsorption. Moreover, the hydrophobic nanoparticles are more efficient in reducing surfactant adsorption than hydrophilic nanoparticles because hydrophobic attractions are typically stronger than hydrogen bonding.[112] Ahmadi and Sheng[114] investigated the effects of hydrophobic and hydrophilic silica nanoparticles on anionic SDS adsorption on carbonate rock sample and observed that hydrophobic and hydrophilic silica nanoparticles could decrease the adsorption loss of the SDS surfactant through hydrophobic interaction and hydrogen bonding, respectively. Wu et al.[107] also reported that silica nanoparticles with a concentration up to 0.3 wt % can effectively reduce the SDS adsorption. However, at a higher concentration (e.g., 0.4 wt %), silica nanoparticles became aggregated and did not reduce surfactant adsorption on the rock surface. In summary, nanoparticles in surfactant solutions can decrease the adsorption of surfactants onto rock surfaces and reduce the loss of surfactants from the solution; therefore, they can increase the efficiency of altering the rock surface wettability.

3.3. Factors that Affect Wettability Alteration. When nanofluid is used in wettability alteration, the stabilities of the nanoparticles in the suspension play critical roles in their efficacy in wettability alteration. Disjoining pressure, the principal mechanism in wettability alteration, comes into play when the nanoparticles form a wedge-film like structure at the reservoir−oil interface and arrange themselves in one, two, and three layers. In the wedge film, the disjoining pressure is higher at the tip, where nanoparticles are arranged in one layer.[10] However, in an unstable nanoparticle suspension, the nanoparticles will be unable to form a single-layer structure due to the agglomerate formation. Therefore, the disjoining pressure will be less effective. Nanoparticles and electrolyte concentrations can affect the stability of the nanofluid. Electrostatic interactions between the nanoparticles and reservoir rock surfaces play a significant role in wettability alteration through the adsorption of nanoparticles on the surface.[17] When a surfactant and nanoparticle mixture are used for wettability alteration, the mechanism and wettability alteration capability depend on the nanoparticle surface wettability (i.e., hydrophobicity and hydrophilicity).[107,112,114] Usually, hydrophobic nanoparticles tend to be more effective in increasing the surfactant and nanoparticle mixture's capability to alter wettability due to the strong hydrophobic bond between the hydrophobic nanoparticle and surfactants' hydrophobic tail.[112,114]

4. INHIBITING ASPHALTENE DEPOSITION

Asphaltene refers to the heaviest and most polar fraction of bitumen and heavy and extra-heavy oils. Asphaltene has a heterogeneous chemical structure, which includes polyaromatic cores attached to aliphatic chains containing heteroatoms (e.g., oxygen, nitrogen, sulfur) and metals (e.g., iron, vanadium, and nickel).[115−117] It is soluble in light aromatic hydrocarbons such as toluene, pyridine, or benzene, but insoluble in low-molecular-weight paraffin such as *n*-heptane or *n*-pentane.[118] Asphaltene might precipitate in reservoirs due to changes in the reservoir conditions, such as temperature, pressure, and oil composition.[119] The deposition of asphaltene can reduce rock permeability and change rock wettability to oil-wet, therefore decreasing the EOR efficiency. In addition, the presence of asphaltene can increase the operating cost because it may plug pipes and processing equipment.[115] Hence, it is very important to remove asphaltene or prevent asphaltene deposition. Prevention methods receive more attention than removal methods due to their long-lasting effects and low costs.[119,120] Several types of prevention methods are available to inhibit asphaltene deposition, including mechanical methods, chemical treatment, thermal treatment, bacteria treatment, and ultrasonic treatment.[119,121] Each of these methods has its disadvantage, such as transience, high cost, and environmental unfriendliness.[119,121] In recent years, nanoparticles have been used as agents to inhibit asphaltene precipitation due to their unique characteristics, including, but not limited to, high surface to volume ratio, surface functionality, and thermal catalytic action that reduce the required temperatures for reactions to take place.[121,122]

4.1. Mechanisms of Nanoparticle-Assisted Asphaltene Deposition Inhibition. Nanoparticles can adsorb asphaltene (Figure 9) so that asphaltene can suspend in reservoir oil instead of depositing onto rock surfaces (Figure 9).[123] In general, asphaltene adsorption on solid surfaces can take place by van der Waals forces, electrostatic interactions, hydrogen bonding, charge transfer, and steric interactions.[124] Adsorption of asphaltene onto nanoparticle surfaces is mainly due to acid−base interactions, electrostatic attraction, and hydrogen bonding.[123] Because the asphaltene is basic, the efficiencies of adsorbing asphaltenes are primarily dependent on the nanoparticle acidity. In addition, higher surface charge

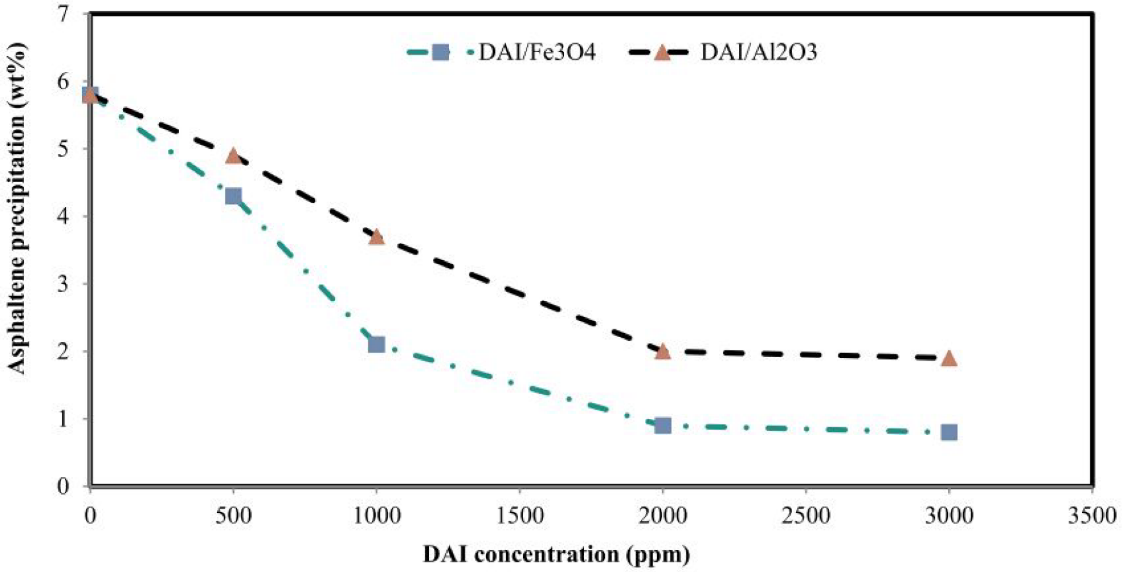

Figure 10. Figure showing the effect of the concentration of direct asphaltene inhibitors (DAI) on asphaltene precipitation. Reproduced with permission from ref 119. Copyright 2020, Elsevier.

density can enhance the stability of the colloidal system and can positively influence the electrostatic interaction depending on the charge type. The isoelectric point (i.e., the pH at which the net charge of the surface is zero) of the asphaltenes are in the range of pH 3−4.5; hence, asphaltene is negatively charged in the neutral condition.[125] Positively charged nanoparticles and asphaltene can then readily form electrostatic bonds.[123] Hosseinpour et al. studied the sorption of asphaltene onto three types of nanoparticles, i.e., acidic (WO_3 and NiO), amphoteric (Fe_2O_3 and ZrO_2), and basic (MgO and $CaCO_3$).[123] They found that the highest adsorption occurred on NiO nanoparticles due to the positive surface charge, high charge density, and acidity. In general, the ultimate adsorption efficiency of a nanoparticle depends on its acidic nature, surface charge density, and surface-to-volume ratio. Shojaati et al.[121] investigated asphaltene adsorption on Fe_3O_4, NiO, and γ-Al_2O_3 metal oxide nanoparticles. They found the highest adsorption of asphaltene on γ-Al_2O_3 nanoparticles due to their acidities and high specific surface areas.[121] Amphoteric particles, or particles that have the ability to act either as an acid or a base, also can adsorb asphaltene via acid−base interactions between the acidic sites of the particle and the basic asphaltene.[119] Azizkhani and Gandomkar[119] studied the adsorption of asphaltene on Fe_3O_4 and Al_2O_3 nanoparticles, both amphoteric, and found higher adsorption on amphoteric Fe_3O_4 nanoparticles, which was attributed to the electrostatic interaction and the higher specific surface area of Fe_3O_4 (286.9 m^2/g) compared to Al_2O_3 (125 m^2/g). When asphaltene adsorbed to basic nanoparticles, it was attributed to the electrostatic interactions between the positively charged particles and negatively charged asphaltene. The asphaltene adsorption efficiency is also dependent on the solution pH because the solution pH determines the surface charges of nanoparticles. For example, TiO_2 nanoparticles are efficient in inhibiting asphaltene precipitation in acidic conditions but not in basic conditions,[126,127] because the TiO_2 nanoparticle has a point of zero charge of 6.8. An increase in the temperature adversely affects the adsorption process because the adsorption is an exothermic process, where energy is released from the system.[115,128,129] Higher pressure also promotes the adsorption of asphaltene as the high pressure increases the tendency to associate asphaltene with nanoparticles.[129] Some studies[119,126] reported that nanoparticles could prevent asphaltene precipitation up to an optimum concentration. Beyond this concentration, an increase in nanoparticle concentration can reduce the adsorption of asphaltene because a concentration increase will result in nanoparticle aggregation, thereby reducing active surface sites.[116,119] For example, Azizkhani and Gandomkar showed that both 500 and 1000 ppm of Fe_3O_4 and Al_2O_3 nanoparticles had higher asphaltene precipitation inhibition capabilities than 2000 and 3000 ppm of Fe_3O_4 and Al_2O_3 nanoparticles, as shown in Figure 10.[119]

4.2. Factors That Affect Inhibiting Asphaltene Deposition by Nanoparticles. Nanoparticles prevent asphaltene deposition through the adsorption of asphaltene on their surfaces. Nanoparticle properties such as surface area to volume ratio, surface functional group, and surface charge density control the mechanism and nanoparticle success in asphaltene deposition prevention. As the asphaltene is basic, the acidic nanoparticles around neutral pH are more effective in asphaltene adsorption.[123,130] The increase in the surface charge density makes the interaction strong. A high surface area to volume ratio will increase the number of adsorption sites, thereby improving asphaltene deposition prevention.[131] Functionalization of the nanoparticle can increase the asphaltene adsorption significantly through the favorable interaction between nanoparticle functional groups and the heteroatoms present in the asphaltene.[132] Nanoparticle porosity can influence the asphaltene adsorption capacity,[132] such as coating the Co_3O_4 nanoparticle with SiO_2 porous nanofilm increased the adsorption capacity.[133]

5. VISCOSITY MODIFICATION

The effectiveness of an EOR process is often measured by sweep efficiency, which refers to the fraction of the reservoir section contacted by the injection fluid. When the injection fluid (e.g., displacing fluid) has a lower viscosity than the displaced fluid (e.g., crude oil), an unstable interface between the two fluids will develop, leaving a significant amount of the displaced fluid behind. This process is referred to as viscous fingering,[12,42] which causes poor sweep efficiency and early breakthrough in the EOR process. High molecular weight, high viscosity, and water-soluble polymer flooding are the most widely used techniques to mitigate viscous fingering and increase sweep efficiency.[31] However, due to chemical, thermal, or biological degradation in porous media, polymers might be degraded so that the viscosity of the injection fluid will be reduced. As a consequence, polymer flooding will not be able to efficiently mitigate the viscous fingering.[134] In addition, in a highly saline condition, polymers may curl up because the charged ions cause a decrease in the electrostatic repulsion force between the polymer molecular chains by creating an electrostatic shielding or reducing the electric double layer potential.[135,136] Hence, the hydrodynamic volume (i.e., the volume of a polymer coil when it is in solution) and the viscosity of the polymer solution decrease.[136] In recent years, nanoparticles have been used in conjunction with polymers as viscosity-modifying agents in displacing fluid.[20,137]

5.1. Polymer and Nanoparticle Mixtures for Viscosity/Mobility Ratio Modifications. Nanoparticles work as thickening agents in polymer solutions by forming a network structure through hydrogen bonding between the nanoparticle surface groups and the polymer structures.[18] If a three-dimensional network structure is formed, it can decrease the mobilities of liquid molecules; hence, the viscosity of the fluid will be increased. Under low shear stress, this 3D network forms, and with the increase in the shear stress, the 3D network breaks, which provides the liquid with pseudoplastic behavior.[18] However, it is important to maintain a balance between the attractive forces and the repulsive forces acting in the suspension to prevent the process of gelation. If the attraction force between the nanoparticles and polymer is too high, the gelation process may start, which is not favorable for the EOR application.[138] Moreover, the gelation process can be controlled by adjusting the nanoparticle size, pH, and salinity of the suspension.[18,139] Zeyghami et al.[18] investigated the effects of hydrophilic fumed silica nanoparticles that got widespread use due to their surface silanol group ability to form network structures through hydrogen bonding in the liquid phase by modifying the viscosity of hydrolyzed polyacrylamide (HPAM) and sulfonated polyacrylamide (SPAM) polymer solutions. They found that the viscosity of the SPAM solution increased with the addition of fumed silica nanoparticles; however, the viscosity of the HPAM solution diminished at low concentrations and increased at high concentrations of fumed silica nanoparticles. This phenomenon was explained by the different interactions between the polymers (i.e., HPAM and SPAM) and fumed silica nanoparticles. Only when nonelectrostatic forces, such as hydrogen bonding, were high enough to compensate for the electrostatic repulsion force could negatively charged polymers adsorb onto negatively charged silica nanoparticles.[140,141] Because the sizes of negatively charged groups on HPAM are similar to the sizes of uncharged amide groups, the repulsive force was not strong enough to hinder the adsorption of the HPAM onto nanoparticles. In addition, the nanoparticle adsorption hindered the stability of the polymer solution by causing curled polymeric chains, which resulted in the polymer's loss from the solution. The adsorption of HPAM onto the nanoparticle probably decreased the previously established hydration effect between the polymer chain and the water molecules.[142] The decrease in the hydration effect caused the curled polymeric chains. A similar phenomenon was observed when carbon quantum dots interacted with the HPAM.[142] As the polymer's concentration in the solution decreased, the viscosity of the solution was reduced. On the other hand, SPAM has much larger negative groups compared to uncharged amide groups, which caused a stronger repulsive force and hindered the adsorption interaction between SPAM and the negatively charged silanol group of the nanoparticles and SPAM. Therefore, the nanoparticle remained in suspension in the SPAM solution and contributed to the thickening effect as it contributed to the thickening effect in a water solution. At the high concentration of fumed silica nanoparticles, the increase in the viscosity of HPAM is probably because the adsorption interaction was not dominant, rather the presence of the nanoparticle in the suspension contributed to the increase in viscosity similar to the SPAM solution.[18] When hydrophilic SiO_2 nanoparticles were added to the partially hydrolyzed polyacrylamide suspension, the SiO_2 nanoparticles increased the suspension viscosity by forming hydrogen bonds.[143] Al-Anssari et al. observed a similar increase in viscosity when they added ZrO_2 nanoparticles in the polyacrylamide polymer suspension.[144] The addition of ZrO_2 in the suspension strengthened the polymeric network and provided improved viscosity even at harsh reservoir conditions such as high temperatures, salinities, and shear forces.[144]

Li et al.[31] investigated the effect of temperature on nanocellulose grafted by 2-acrylamido-2-methylpropanesulfonic acid and hydrophobic groups (NC-KYSS). They found an increase in the viscosity when the temperature increased from 25 to 50 °C. At low temperature, hydrogen bonding formed around the hydrophobic part of the NC-KYSS, which made the hydrophobic polymers soluble in water.[145−147] However, with an increase in the temperature, the hydrogen bonding broke, which facilitated the hydrophobic interaction among the polymers.[148,149] The increase in viscosity with the increase in temperature is probably due to the network formed from the hydrophobic interaction between the polymers. The viscosity then decreased when the temperature further rose above 50 °C (Figure 11). A temperature higher than 50 °C

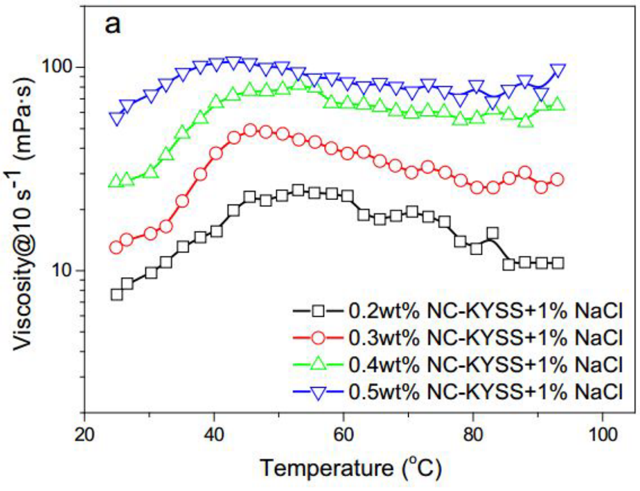

Figure 11. Effect of temperature on the viscosity of NC-KYSS nanofluid, (NC-KYSS refers to the nanocellulose grafted by 2-acrylamido-2-methylpropanesulfonic acid and hydrophobic groups). Reproduced with permission from ref 31. Copyright 2017, Elsevier.

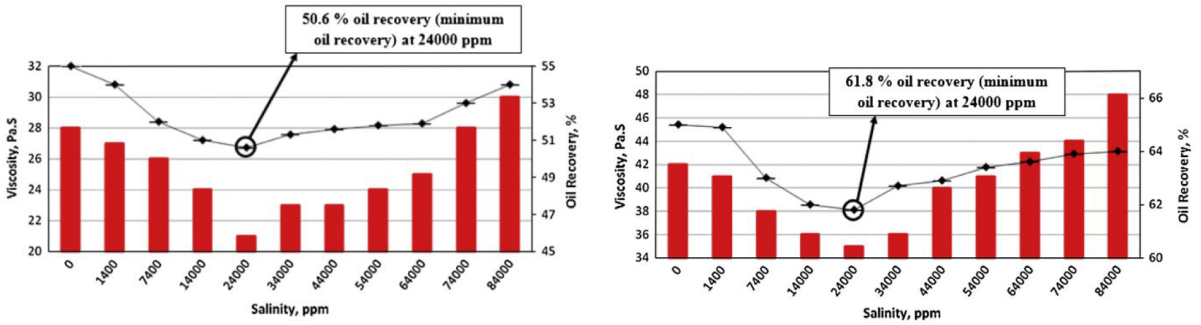

Figure 12. Effect of salinity in viscosity and oil recovery by polymer flooding in the absence (left) and presence (right) of silica nanoparticles. Reproduced with permission from ref 150. Copyright 2014, Elsevier.

probably ruptured the previously formed network due to the increased motion of the NC-KYSS and water molecules, which resulted in a decrease in viscosity.

Maghzi et al.[150] studied the effect of salinity on polyacrylamide flooding and found viscosity and oil recovery decreased when salinity increased from 0 to 24,000 ppm (Figure 12). Multiple charges of polymers surrounded along their chains make them sensitive to the salinity condition. In a low saline condition, an electric double layer formed around the polymer chain, which prevented the polymer from curling up. However, with the increase of salinity, the thickness of the double layer decreased, which onset the polymer's contraction and decreased the solution's viscosity.[151] A further increase in salinity beyond 24,000 ppm increased viscosity and oil recovery.[150] This increase in the viscosity can be explained by the increase in the polarity of the solution with an increase in the salinity. The increased polarity weakened the hydration layer which increased the hydrodynamic volume of the polymer and increased the viscosity.[152] However, in all salinity conditions, silica nanoparticles, in conjunction with the polyacrylamide, increased the viscosity and the oil recovery factor due to the ion−dipole interaction between the cations and silica nanoparticles.[150]

5.2. Factors That Affect Viscosity Modification.

Nanoparticles increase the viscosities of polymeric solutions by forming a pseudopolymeric network through hydrogen bonding.[18,144] When a polymer can provide a sufficient electrostatic force to prevent the nanoparticle entanglement in the polymer chain, the formation of a network structure is possible.[18] An increase in the temperature can rupture the polymeric network and reduce the viscosity.[31] However, an increase in the salinity can contribute to the viscosity increase through its charge screening process.[153] The charge screening leads to the aggregation and formation of large nanoparticles, thereby increasing the solution's viscosity.[153]

6. EMULSION STABILIZATION

During EOR operations, emulsions can block highly pervious cavities, direct flow toward unswept zones,[154,155] decrease mobility,[156] and improve the sweep efficiency.[157−159] Therefore, emulsions are a preferable choice over waterflooding for EOR applications.[156] Both nanoemulsions[160] and microemulsions[161] have been studied for their potentials as field-scale EOR agents. Nanoemulsions, with droplet diameters less than 100 nm, were reported as preferable for small cavity reservoir applications due to their high penetrating ability, excellent dispersibility, and lack of shear thickening and sedimentation.[7,47,162] Harsh reservoir conditions, such as high temperatures (70 °C to more than 120 °C),[163] high salinity (total dissolved solids of approximately 200,000 mg/L),[163] and high pressures, however, can undermine the properties of traditional emulsions (i.e., nano and microemulsions), leading to sedimentation or unexpected phase separation.[12,163] It is essential to form a stabilized emulsion to emulsify and mobilize the residual oil in the reservoir and increase the recovery rate.[12] In recent years, nanoparticles, surfactants, or composites of surfactants and nanoparticles have been used to stabilize emulsions so that they work well in harsh reservoir environments.[16,164,165] It was reported that neither nanoparticles nor surfactants were good emulsifiers when used separately. Nanoparticles do not work well at pH values far from their isoelectric points.[166] When surfactants are used alone, it is difficult to control the emulsion mobility.[7] However, emulsions formed using the composites of nanoparticles and surfactants remain stable at both high and low pH values.[21,166−168]

6.1. Mechanisms Regulating Nanoparticle- or Surfactant-Induced Emulsification.

Emulsifiers' (i.e., nanoparticles, surfactants, or surfactant-modified nanoparticles) properties, such as hydrophobicity and molecular structure, play essential roles in the emulsification process.[169,170] Hydrophilic emulsifiers, including hydrophilic nanoparticles, prefer to form oil-in-water (o/w) emulsions and tend to adsorb at the oil−water interface. Because hydrophilic nanoparticles tend to reside in water, water becomes the dominating phase and forms o/w emulsion. Hydrophilic surfactants will also form o/w emulsions. Because the sizes of the charged headgroups of hydrophilic surfactants are larger than the alkyl chain, these charged head groups tend to disperse in water, making the water phase dominant and leading to the o/w emulsion. On the other hand, hydrophobic emulsifiers prefer to form water-in-oil (w/o) emulsion.[77,171,172] The spherical hydrophobic particles remain in the oil phase so that the oil phase becomes dominant. The adsorption of hydrophobic nanoparticles at the interface leads to the formation of w/o emulsions. When hydrophobic surfactants are used as emulsifiers, their alkyl chains, which are larger than the charged headgroup, tends to reside in the oil phase, making the oil phase dominant and leading to the formation of w/o emulsions. The stability of the emulsion depends on the adsorption energies of the emulsifiers at the oil−water interface.[77] For strongly hydrophobic or hydrophilic emulsifiers, strong repulsions exist among hydrophobic or hydrophilic emulsifiers at the interface, which lead to low adsorption energy[173] and unstable emulsions.[162] In order to form a stable emulsion, nanoparticles need to be either moderately hydro-

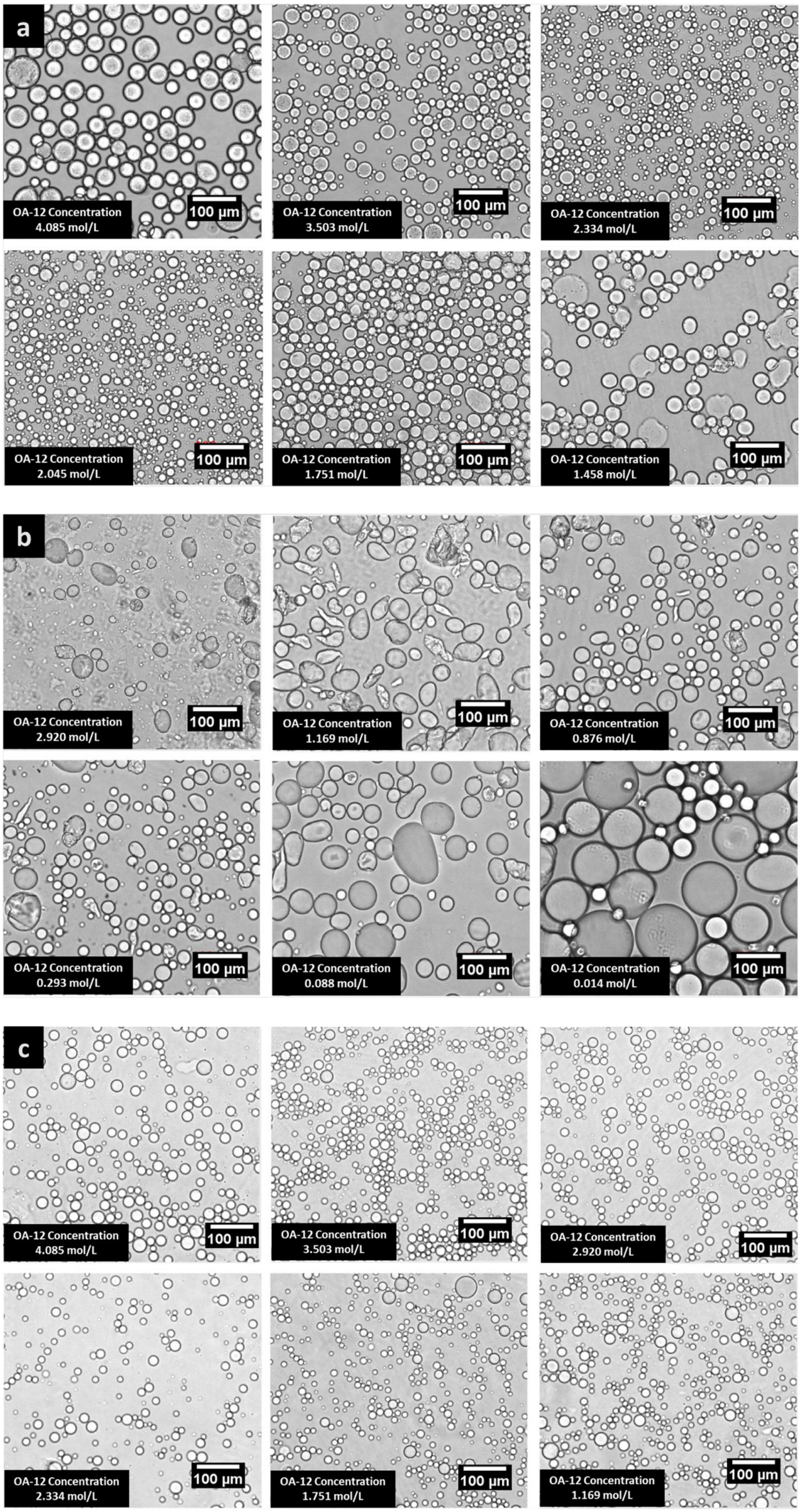

Figure 13. Optical microscopy images of (a) w/o (water-in-oil) emulsions of different dimethyl dodecyl amine oxide (OA-12) concentrations (i.e., 4.085, 3.503, 2.334, 2.045, 1.751, and 1.458 mol/L) with silica nanoparticles and (b) o/w (oil-in-water) emulsions of OA-12 with silica nanoparticles. The concentrations of OA-12 were varied by 2.920, 1.169, 0.876, 0.293, 0.088, and 0.014 mol/L, while silica nanoparticle

Figure 13. continued

concentrations are constant and (c) OA-12 only emulsions (oil-in-water emulsions) at different concentrations, i.e., 4.085, 3.503, 2.920, 2.334, 1.751, and 1.169 mol/L. Reproduced with permission from ref 21. Copyright 2018, Elsevier.

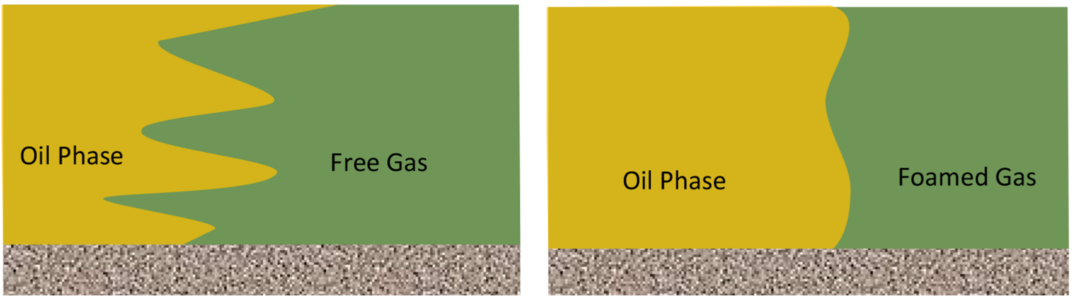

Figure 14. Schematic of gas flooding (left) vs foam flooding (right). Foam flooding reduces viscous fingering by modifying the mobility ratio. Schematic was drawn with the idea from ref 181.

phobic or moderately hydrophilic.[162] The adsorption energy at the interface is dependent on the contact angle of the nanoparticle with the oil−water interface.[162] Usually, a contact angle of 90° leads to the most stable emulsions, and a particle of intermediate hydrophobicity has the property to form a contact angle of 90°.[22,162]

Emulsion stability is also influenced by the droplet size and the viscosity of the dominant liquid phase,[162,167,173] both of which are related to the concentration of the emulsifier.[167] When nanoparticles are used as emulsifiers, the droplet size decreases with the increase of the nanoparticle concentration up to a specific limit.[167] The high concentration results in the formation of a tightly packed film which prevents the demulsification mechanism and provides long-term emulsion stability.[103] Moreover, the increase in concentration reduces the interfacial tension and then decreases the droplet size.[170] Once a certain threshold is reached, nanoparticle concentration no longer influences emulsion droplet size, even though concentration plays an important role to prevent sedimentation or creaming.[170] Particularly, excessive nanoparticles can increase liquid viscosity, which could reduce the tendency of emulsion droplets moving up to form a separate layer on top of the continuous phase (i.e., creaming[174]) or moving down (i.e., sedimentation). Similar to emulsions stabilized by nanoparticles, the droplet size of the emulsion stabilized by a surfactant decreases until a critical surfactant micelle concentration reaches.[166] Afterward, the increased concentration increases the viscosity and density of the liquid.[166,174] At the critical micelle concentration, surfactants adsorb around emulsion droplets and can prevent them from coalescence for a long period of time due to both a small droplet size and a favorably thickened dispersed phase.[166]

6.2. Mechanism Involved in Composite of Nanoparticle and Surfactant-Induced Emulsifications. Nanoparticles and surfactants have been used together to explore their synergistic effects on emulsion stabilization.[21,166,167] The emulsion type, i.e., o/w or w/o, depends on the hydrophilic−lipophilic balance number (Figure 13).[175] The stability of the emulsion also relies on the hydrophobicity of the nanoparticle−surfactant composite.[166] Although highly hydrophilic nanoparticles are poor emulsifiers if used alone, they become excellent emulsifiers when mixed with cationic or anionic surfactants and are capable of preventing coalescence, sedimentation, or creaming.[166,167] If nanoparticles are negatively charged, the addition of positively charged surfactants can modify their hydrophobicities by creating monomers on nanoparticle surfaces.[21,166,167] Due to electrostatic attraction, heads of surfactants cling to surfaces of nanoparticles, and the charge-neutral alkyl chains remain floating in the liquid phase. With more surfactants on nanoparticle surfaces, the hydrophilic nature of the nanoparticle decreases while its hydrophobicity enhances. When the adsorbed surfactants saturate the monolayer, a double-layer formation is initiated by chain−chain interactions.[21] The alkyl chains of the attached surfactants float from nanoparticles, which attract alkyl chains of excessive surfactants in the aqueous phase due to chain−chain interactions and then leave charged head groups outside. As charged head groups remain exposed, they bring back the hydrophilic nature of the composite. When nanoparticle and surfactant composites shift from hydrophilic to hydrophobic and vice versa, the emulsion type also changes.[21,166] For positively charged nanoparticles, the addition of a negatively charged surfactant also shows a similar mechanism. The maximum stability of the emulsion has been observed when the adsorption of surfactants on nanoparticles modifies the wettability enough to induce flocculation of the nanoparticle.[166] The nanoparticle and surfactant composite prevents emulsion coalescence by adsorbing at the interfaces and also prevents creaming by increasing the viscosity of the solution.

6.3. Factors That Affect the Emulsion Stabilization Process. Electrolyte concentration, as well as pH, changes the surface potential of the nanoparticle, which may lead to aggregation, thereby influencing the stability of the emulsion.[170] Nanoparticles tend to self-aggregate when pH is close to the isoelectric point, which is pH value at which the surface potentials of nanoparticles is close to zero. Therefore, at the isoelectric point of nanoparticles, the emulsion is destabilized since the aggregated nanoparticles are weak stabilizers. When the pH value is far from the isoelectric point, nanoparticles are strongly hydrophilic or hydrophobic, which is not suitable for a stable emulsion.[170] Moreover, the nanoparticle surface wettability controls the emulsion droplet size distribution over time and emulsion stability. Partially hydrophobic nanoparticles provide long-term stability compared to hydrophobic nanoparticles.[176] The sizes of nanoparticles also affect the foam stability, such as the smaller sizes of nanoparticles

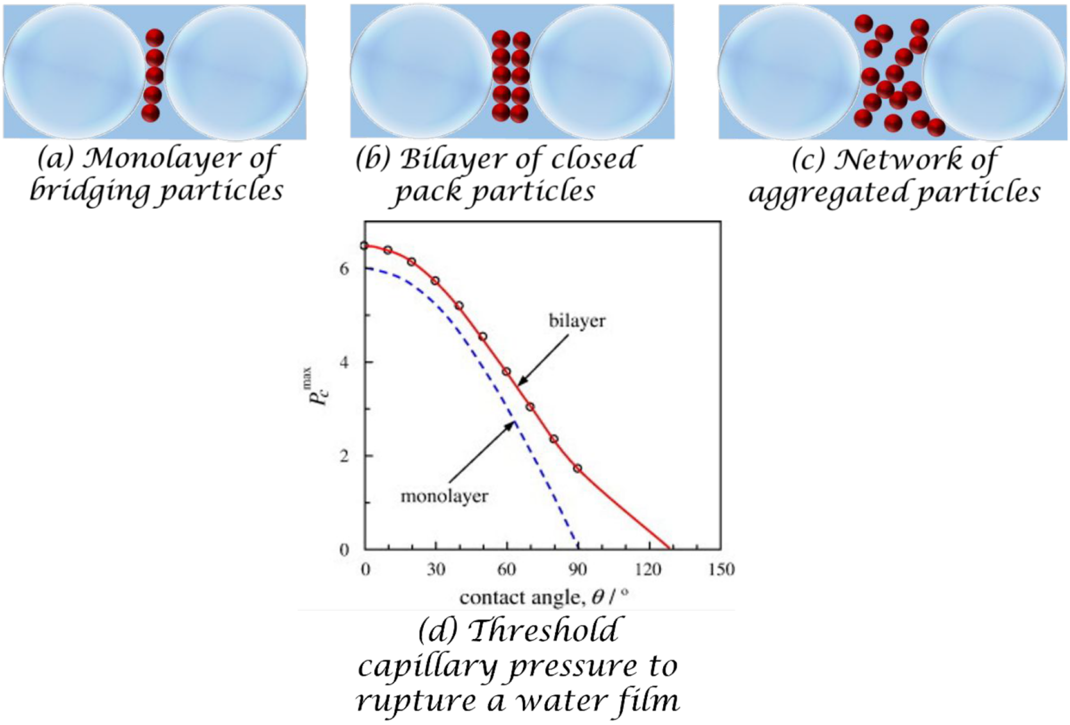

Figure 15. Particle arrangement at the bubble−liquid interface during the nanoparticle foam stabilization process: (a) monolayer bridging of the particles, (b) bilayer closed pack particles, (c) network of the particles aggregate (schematic drawn with the idea from ref 186), and (d) threshold capillary pressure to rupture a water film stabilized by a spherical particle at a different contact angle. Reproduced with permission from ref 26. Copyright 2008, Elsevier.

proved to be more efficient in providing foam stability compared to larger-sized nanoparticles.[177]

7. FOAM STABILIZATION

Gas flooding, which involves the injection of carbon dioxide, nitrogen, or other hydrocarbon gases into reservoirs, accounts for about 39% of the global contribution from EOR production.[49,178,179] Depending on whether injected gas can thoroughly mix with oil, there are two types of gas flooding technologies, including immiscible gas flooding and miscible gas flooding. Gas flooding can swell the oil, reduce the oil viscosity, and decrease the interfacial tension, thereby increasing the EOR efficiency.[180] Compressed gas is an excellent choice as a displacing fluid in the EOR application because it can maintain a high density over a range of high temperatures and pressures, has extremely low solubility in water, and is cost effective. However, there are challenges associated with gas flooding, including channeling, viscous fingering, and gravity override.[181,182] Channeling refers to the flow of gas through the highly permeable flow paths in heterogeneous reservoirs. Viscous fingering refers to the unstable interface between two fluids when a less viscous gas replaces a more viscous crude oil. Gravity override refers to the phenomenon where a low-density gas flows on top of denser crude oil, which leads to limited mixing. In addition to addressing these challenges associated with gas flooding (Figure 14), foams also can decrease the amount of solution required to sweep a heterogeneous reservoir, because foams can divert flow from the thicker fracture to a thinner fracture.[182]

Foam is the dispersion of gas in a continuous liquid phase. Gas bubbles are enclosed in cells, and cell walls act like planar, which is called lamellae or bubble film.[183] The efficiency of foam flooding depends on the stability of the foam, its ability to prevent liquid drainage, the stabilities of foam films, and the capacity to prevent interbubble gas diffusion.[184] Surfactants, nanoparticles, or surface-modified nanoparticles are commonly used to improve the capability of generating foam (i.e., foamability) and foam stability (i.e., dynamics of foam height and volume change with the time immediately after generation).[185] Generally, nanoparticles modified with surfactants or functional groups perform better than surfactants or nanoparticles alone in improving foamability and foam stability.[184,186]

7.1. Mechanisms Involved in Foam Stabilization.

Nanoparticle hydrophobicity plays a crucial role in foam stabilization.[187] For example, foam stabilized by particles with contact angles of slightly less or more than 90° is very stable, which can be explained by the adsorption free energy.[74,173,187] The particle adsorption energy (E_d) at the interface can be estimated as[162]

$$E_d = \pi R^2 \gamma_{aw} (1 - |\cos\theta|)^2 \quad (3)$$

where R is the nanoparticle radius, γ_{aw} the interfacial tension at the air−water interface, and θ the contact angle. According to eq 3, the value of E_d is very low for highly hydrophilic (contact angle < 30°) or highly hydrophobic (contact angle > 150°) particles, which cannot provide high foam stability.[186] On the other hand, a few nanometer-sized particles (i.e., 10 nm) with contact angles close to 90° can achieve an E_d value on the order of magnitude of $10^3 kT$ (k = Boltzmann constant, and T = temperature).[186] A high E_d value implies that particles can attach to the surface irreversibly and provides ultrastability to the foam.[186,187]

Nanoparticles arrange in the bubble−liquid interface as a bridging monolayer, a closed-packed bilayer, or a network of aggregated multilayers (Figure 15).[26,186] The particle arrangement at the foam film influences the maximum capillary pressure that foam films can withstand before rupture and coalescence.[26,188] Kaptay[188] has derived eq 4 to quantify the threshold capillary pressure($p_c^{\max}$)

$$P_c^{\max} = \beta \frac{2\gamma_{aw}}{R}(\cos\theta \pm z) \tag{4}$$

where β and z are functions of particle arrangement. Particularly, z is zero for the single-layer arrangement, and $z = 0.633$ for a double-layer particle arrangement when $\theta > 90°$. A decrease in the particle radius (R) suggests an increase in the maximum capillary pressure a film can withstand. Based on this equation, for a monolayer of nanoparticles with a contact angle of 90° or higher, the capillary pressure needed to rupture the film becomes zero. Therefore, particles with a contact angle of 90° or greater are not capable of stabilizing foam by forming a single layer.[26,188−190] However, the particles forming a bilayer at the water film are capable of stabilizing foam up to a contact angle of 129° (Figure 15).[26,188,191]

Nanoparticles and surfactants can be used in synergy to modify the wettability of the nanoparticle to slightly hydrophobic to transfer the nanoparticle from the bulk phase to the gas−liquid interface.[56,192,193] Surfactants usually can get adsorbed on nanoparticle surfaces through the electrostatic attraction force or hydrophobic interaction.[166,167,194,195] At low surfactant concentration (i.e., lower than the CMC), the cationic surfactants get adsorbed in the negatively charged nanoparticle, or the anionic surfactants can get adsorbed in the positively charged nanoparticles through electrostatic interaction.[166,195] Through the adsorption of surfactants, a monolayer is formed on the nanoparticle surface, and the nanoparticle surface wettability becomes slightly hydrophobic due to the outward direction of the alkyl chain from the nanoparticle surface.[195] After the monolayer formation, an increase in the surfactant concentration will initiate a hydrophobic interaction between the adsorbed and floating surfactant tail and change the nanoparticle wettability back to hydrophilic, which is unsuitable for the foam stabilization application.[195,196] However, when nanoparticles and surfactants are used together for foam stabilization, the concentration ratios between nanoparticles and surfactants should be balanced. Suppose the surfactant concentration is not high enough to modify all the particles into slightly hydrophobic. In that case, the remaining hydrophilic particles will be in the suspension and contribute to the viscosity increase of the suspension, decreasing the foamability.[195,197] At too high surfactant concentration, all the particles will be modified back to hydrophilic, increasing the viscosity and eventually decreasing foamability.[195,197] Although the presence of hydrophilic nanoparticles decrease the foamability, it was reported that the formation of a 3D network in the bulk phase by hydrophilic nanoparticles can increase the foam stability.[198] Moreover, a complete monolayer coverage of the surfactant through electrostatic interaction will make the nanoparticle too hydrophobic to stabilize the foam.[192] It was reported that slightly hydrophobic nanoparticles with partial coverages are capable to provide the best foam stability.[192,199−201]

7.2. Factors Affecting Foam Stabilization. When a surfactant is used together with nanoparticles, the surfactant type is important for foam stabilization. For example, Cui et al. showed that positively charged $CaCO_3$ nanoparticles can be surface activated by anionic surfactants and provide high foamability and foam stability.[187] The electrostatic interaction between positively charged $CaCO_3$ and a negatively charged headgroup of surfactants results in monolayer adsorption and surface activation. However, these same $CaCO_3$ nanoparticles cannot be surface activated in the presence of cationic or nonionic surfactants due to the absence of electrostatic interactions between the nanoparticles and surfactants.[187] A similar influence of electrostatic interaction in the foamability and foam stability was observed when negatively charged SiO_2 nanoparticles were used with cationic CTAB.[202] Moreover, the electrostatic repulsion between the nanoparticle and surfactant may decrease the foam stability due to the disorderly arrangement of the surfactant at the interface.[201] The types and concentrations of electrolytes in the solution also influence the properties of the foam, including bubble size, foamability, and foam stability.[25] Additionally, an increase in the temperature can decrease the foam stability and increase its mobility.[25]

8. SUMMARY AND OPPORTUNITIES

The application of nanoparticles for EOR is still at the emerging stage, and most existing studies are at the laboratory scale.[29,32,203,204] The successful application of nanoparticles in EOR in the field is dependent on an advanced understanding of the mechanisms that regulate the fundamental processes in nanoparticle-induced EOR. We comprehensively reviewed fundamental mechanisms associated with nanoparticle-assisted EOR, including wettability alteration, IFT reduction, mobility ratio enhancement, emulsion stabilization, and foam stabilization. For each mechanism, we discussed how various factors, including nanoparticles' size and concentration, pH condition, nanoparticles' isoelectric point, salinity condition, temperature, and reservoir pressure, can affect how nanoparticles regulate each mechanism. Below is a list of opportunities that we identified for further investigation:

- Both hydrophobic and hydrophilic nanoparticles are proven efficient in IFT reduction. Both positively and negatively charged nanoparticles are used together with surfactants to enhance EOR. Systematically comparing the performances of nanoparticles with different hydrophobicity levels and surface charges can provide deep insights into the selection of nanoparticles for industry-scale EOR applications.
- Nanoparticle adsorption at the interface can enhance EOR by reducing IFT and altering rock surface wettability. However, the sorption of nanoparticles can reduce the nanoparticle concentration in the solution, which can minimize the EOR efficiency. Disjoining pressure resulting from nanofluid flooding is effective in altering the surface wetting properties while maintaining nanoparticle concentration in the bulk solution. A comprehensive study to investigate the optimal conditions to utilize the combined effects of disjoining pressure and nanoparticle adsorption is needed.
- The stabilities of nanofluids are essential to prohibit asphaltene deposition in the reservoir. Positively charged nanoparticles have been proven effective in maintaining stable nanofluids and preventing asphaltene deposition. Systematic studies are needed to guide the formation of positively charged stable nanofluids to prevent asphal-

tene deposition. The isoelectric point of the nanoparticle plays an essential role in the surface charge density and stability of the nanoparticle in the suspension. However, the current scarcity of data availability makes it difficult to establish a relationship between the isoelectric point and the efficiencies of nanoparticle in asphaltene deposition prevention. A systematic study is required to provide a relationship between the isoelectric point of the nanoparticle and the efficiency of asphaltene deposition prevention.

- Nanoparticles in conjunction with polymers have been proven efficient in modifying the mobility ratio between the displacing fluid and displaced oil. During the operation, polymers can be lost from the solution. Also, the use of polymers can increase the cost and may have negative environmental impacts. Additional studies are needed to understand how the increase of the nanoparticle concentration alone can contribute to mobility ratio improvement.
- Studies have proven that nanoparticle hydrophobicity has an essential impact on their abilities to stabilize emulsions. Quantitative studies on the relationship between nanoparticles' hydrophobicities and emulsions' stabilities are needed to enhance industrial-scale applications.
- The arrangement of nanoparticles at the bubble−liquid interface plays a crucial role in foam stabilization. It is still not well understood what characteristics of nanoparticles affect their arrangements, which require additional research.

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Notes

The authors declare no competing financial interest.

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