

# Application of Surfactants in Hydraulic Fracturing for Enhanced Oil/Gas Recovery

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## Abstract

Surfactants are widely used in the hydraulic fracturing process to enhance oil/gas productivity and reduce energy consumption by controlling for optimal viscosity of fracturing fluids, reducing surface/interfacial tension between the shale formations and the fracturing fluid, assisting fluid recovery after fracturing, altering the wettability of rock and reducing flow friction of fracturing.

The up-to-date progress of the application of surfactants in fracturing fluids for enhancing oil/gas recovery are summarized comprehensively from the aspects of single surfactant system, mixed surfactant system, surfactant combination with nanoparticles and polymers, mainly concentrating on the effect of surface tension reduction, wettability alteration and drag reduction from the point of experimental research and numerical simulation. And the broad application prospects of surfactant compound with nanoparticle and polymer are also outlooked.

**Keywords:** Surfactant; Oil/gas recovery; Surface tension; Drag reduction; Wettability alteration

## Introduction

Since its discovery, oil and gas has remained the world's major energy source despite the recent contribution from renewable energy [1]. Hydraulic fracturing is normally applied to stimulate low/ultralow permeability reservoirs in order to obtain an economical hydrocarbon production rate [2]. However, 50-95% of the injected fluid is typically reported to remain in the formation due to the capillary effect [3,4], which will hinder hydrocarbon production and cause formation damage [5]. The capillary force, one form of capillarity, could be mainly responsible for the surface tension of fracturing fluid [6]. On the other hand, gas exploited from reservoirs is prone to condense into liquid under pressure conditions below the saturation point [7], then a liquid phase, known as condensate bank is formed and retained by capillary forces in the liquid-wettable rock, reducing the gas permeability and thus the wellbore productivity [8], especially for low-permeability reservoirs and tight reservoirs [9]. Chemical enhanced oil/gas recovery has been proven to be a promising method to recover the trapped gas by reducing the surface tension and altering the wettability of rock surface from liquid to gas by use of specific surfactant [10]. Several surfactant-based fracturing fluid additives have been developed in an attempt to enhance the flow back and oil/gas production [11-13]. Simultaneously, when the concentration of surfactant in the fracturing fluid reach to some extent, worm-like micelle will form, thus reducing the flow resistance of fracturing fluid [14]. In recent years, a viscosity surfactant-based clear fracturing fluid has been applied as the substitute of traditional polymer-based fracturing fluids with characteristics of high flow back rate, high productivity, low friction and low formation damage [15].

Surfactants, one of the most important additive in fracturing fluid, are used to control for optimal viscosity of fracturing fluids, reduce surface tension between the shale formations and the fracturing fluid, assist fluid recovery after fracturing, alter the wettability of rock and reduce flow friction of fracturing [16]. In some instances, surfactants may also act as biocides or clay stabilizers [17]. The role of surfactants mentioned above are all helpful to enhance oil/gas productivity and reduce energy consumption [18].

The present review about the application of surfactants in fracturing fluid are concentrating on the recent trends and reach progress of compounds of different surfactants [19] and single special surfactants such as polymeric surfactants [20], Gemini surfactants [21] and so on. However, to the best of our knowledge, there is very limited of such a review that comprehensively introduce the various and latest applications of different surfactants compound with different kinds of chemical additives like nanoparticle, polymer to enhance the productivity of oil/gas from low permeability reservoirs, domestically and abroad so far. In the following sections, we will summarize and discuss the up-to-date progress of the application of surfactants in fracturing fluids for enhancing oil/gas production comprehensively, from the point of the interaction of different kinds of surfactants as well as surfactants with nanoparticle and polymer, mainly concentrating on the effect of surface tension reduction, wettability alteration and drag reduction, from the point of experimental research and numerical simulation. The review is expected to be of references for the researchers working in the field.

## Single Surfactant System

### Traditional surfactant

**Experimental study of traditional single surfactant:** Typical traditional surfactant such as anionic surfactant sodium dodecyl sulfate (SDS) and cationic surfactant cetyltrimethylammonium bromide (CTAB), cetyltrimethylammonium chloride (CTAC) can reduce

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surface tension to some extent around critical micelle concentration (CMC). The effects of SDS and CTAB on the interfacial tension (IFT), wettability alteration and spontaneous imbibition of oil and outcrop rock samples obtained from Aghajari reservoir were investigated. The experimental results show the obvious effect of these surfactants at very dilute concentrations [22]. Mirna et al. studied the performance of the cationic surfactant CTAC on the enhancement of the oil recovery factor from heavy oil-impregnated calcite cores. The interaction energies between CTAC and either of some molecules present in oil were theoretically determined within the framework of the Density Functional Theory and experimentally analysed respectively [23].

Mohammad et al. investigate adsorption kinetics and equilibrium of a novel nonionic surfactant *Glycyrrhiza Glabra* in aqueous solutions for EOR and reservoir stimulation purposes [24]. The result shows that the surfactants trend to adsorb onto carbonate, which can be used for enhancing oil recovery from carbonates effectively and economically [25]. Amit et al. synthesize a carboxybetaine based zwitterionic surfactant by quaternization of dodecyltrimethylamine with chloroacetate, which is characterized by FTIR, <sup>1</sup>HNMR and TGA analysis. The synthesized surfactant is effective in decreasing the IFT between crude oil and water at a low CMC and changing the wettability of oil wet quartz sample to water wet. Sand pack experiments showed additional oil recovery of 27.03% by flooding of surfactant slug at CMC, which evidence the suitability of the synthesized zwitterionic surfactant for application in chemical EOR. Nilanjan et al. synthesize a novel anionic surfactant from coconut oil as an alternative during oil/gas recovery, which exhibits good salt tolerance levels and long-term thermal stability, as well as lower cost [26].

**Numerical simulation of traditional single surfactant:** With the development of computer science and technology, people have not satisfied with the physical and chemical properties of substances obtained by experimental methods. Computer simulation studies and experimental studies can support and supplement each other. Experimental studies provide experimental data that is essential for computer simulation, while computer simulations provide data that cannot or difficult to acquire by experiment [27]. Molecular dynamics simulation and molecular fragment dynamics simulation are helpful to understand the self-assembling behaviour and dynamic characteristics of surfactants, which have become the research hotspot of surfactant numerical simulation.

Chanda et al. simulate the surface coverage, surfactant orientation and kinetic details of a single layer of nonionic surfactant C12E2 adsorbed at the air/water interface at CMC and indicates that the surfactant has stronger mobility on the interface plane than in the normal direction of the interface [28]. The hydrophilic head exhibits faster reorientation dynamics due to the shorter hydrophobic tail length and tends to tilt toward the interface plane.

Pang et al. perform molecular dynamics simulations absorption of the surfactants containing the same hydrophobic carbon: anionic surfactant SDS, cationic surfactant dodecyl trimethyl ammonium bromide (DTAB), nonionic surfactant octaethylene glycol lauryl ether (C12E8) and surface activator siloxane modified by oxyethylene and oxypropylene (DSEP) at the air/water interface [29]. The results show that the head groups of SDS and DTAB are entirely hydrated and located in the region of water, not all the oxyethylene groups of C12E8 immerse in the water layer. Due to the large bond angle and long bond length, the Si-O bond can freely rotate and tilt. Therefore, the DESP chain is flexible and spreads easily at the interface, showing a stronger surface activity and increasing the solubility of the remaining surfactant. In the

presence of DESP, the SDS monolayer is relatively ordered and compact, and some of the DTAB molecules are in water while the others at the interface are organized, whereas the C12E8 monolayer turns out to be disordered and loose. Protonated oxyethylene group possesses weak positive charge in aqueous solutions.

There is the electrostatic attraction between DESP and SDS, so the electrostatic repulsion between headgroups of SDS can be partially screened. As a result, the monolayers are more closely arranged in the presence of DESP. Meanwhile, there is a weaker electrostatic repulsion between DESP and DTAB. Additionally, DESP, having higher surface-activity, is prone to extend at the surface. Consequently, some DTAB molecules dissolve in water, and the others at the surface are well ordered, far apart from each other. DESP mainly depends on Vander waals and hydrophobic interactions to affect the properties of the C12E8 monolayer. Only the oxyethylene groups of C12E8 close to the surface can interact with DESP by hydrogen bond. The simulation results confirm that the aggregation behaviors of hydrocarbon surfactants at the air/water surface are obviously influenced by a siloxane surfactant, which can provide a theoretical fundament to predict the properties of surfactant mixtures.

Truszkowski A use molecular fragment dynamics simulation to study the behaviour of non-ionic surfactant polyoxyethylene alkyl ether at the water-air interface [30]. The calculation results show that the longer the carbon chain of the surfactant, the lower the CMC, a longer carbon chain leads to a lower surface tension under the condition of same concentration below CMC. The surface tension tends to fluctuate when the CMC is reached. At the same time, the number of surfactant molecules at the water-air interface per nm<sup>2</sup> is analysed, the number of surfactant molecules at the interface increases linearly with increasing concentration until the saturated concentration reached. A longer carbon chain leads to a lower concentration at saturation, which is consistent with experimental results.

### Special surfactants

Traditional surfactants have limited ability to reduce surface tension. Liang et al. investigate the effect of different surfactants on reducing the damage resulting from water block by reducing the interfacial/surface tension and enhancing permeability [31]. The result show that surfactants providing moderate reductions on interfacial tension has negative impacts on solving the water block, while surfactants providing the ultralow interfacial tension can enhance matrix permeability and hydrocarbon recovery because of the elimination of the matrix-fracture interaction. In order to obtain ultra-low surface tension, special surfactants are developed.

Fluorocarbon surfactant is one of the most important surfactants which has the highest surface activity, good heat-resistant and salt resistance. However, the synthesis is very difficult and the cost is high, so it is commonly used in combination with cheaper hydrocarbon surfactant [32]. Whereas, fluorocarbon surfactant has poor low temperature resistance and weak affinity with most organic solvents, which limits their applications. Therefore, fluorosilicone surfactant has been developed on the basis of fluorocarbon surfactants [33].

Gemini surfactant encompasses a relatively new generation of surfactants that has unlimited scope in terms of flexibility and economic effectiveness [34,35]. The presence of two hydrophilic head groups and two hydrophobic tails confer unique properties that are well-suited to requirements of different industries [36]. Soheila et al. investigate the aggregation behavior, physicochemical properties and morphology of

the surfactants of ester-containing cationic Gemini surfactants, dodecyl esterquat and dodecyl betainate Geminis in the absence and presence of NaBr electrolyte [37]. The results show that the position of ester bonds in alkyl tail has a considerable effect on both physicochemical properties and aggregation behavior of the ester-containing Gemini surfactants and the salt addition induced the growth of micelles. Palet et al. investigate the equilibrium and dynamic interfacial properties of Gemini surfactants with different spacer lengths and demonstrate declining trend of the interfacial tension with the increase of spacer lengths [38]. Then the equilibrium adsorption characteristics of Gemini surfactant molecules at the oil-aqueous interface are explained using the Langmuir and Freundlich isotherms, and conclude that Gemini surfactants molecules at low concentration from the bulk to the interface may be described as a diffusion controlled process, but is best described as a combination of adsorption and diffusion controlled adsorption at higher concentration, till the interface is completely saturated.

Polymeric surfactant represents a very attractive alternative to chemical enhanced oil/gas recovery, because they can provide simultaneously increase in water viscosity and decrease in interfacial tension, both beneficial for the efficiency of the fracturing process [39]. However, the synthesis are often challenging and costly, Patrizaio et al. suggest a much more promising way by the introduction of biopolymers and bio-based monomers [40,41].

## Mixed Surfactants System

### Experimental study of mixture surfactants

Although a single surfactant can reduce the surface tension of the solution, its ability to form micelles is weak, so it is necessary to compound multiple chemical agents to acquire better performance at lower concentration when improving oil/gas recovery. Mixed surfactant systems have gained great interest because of polymorphism of self-assembly structures accessible through simple tuning of composition that have combined properties of the various surfactants in the mixture [42,43].

When ionic surfactant adsorbs on the interface, the strong electrostatic repulsion resulting from the same charged group facilitates the molecular arrangement not too tight. The non-ionic or zwitterionic surfactant will be easier to enter the loose ionic surfactant adsorption layer due to the hydrophobic effect and the possible dipole-dipole interaction. Thus, the electrostatic repulsion will be weakened and the molecules will arrange closely, then the surface activity will be improved [44,45].

It is reported that zwitterionic-anionic mixed surfactant systems offer synergistic interactions in aqueous medium due to polymorphism of self-assembly structures that have combined properties of surfactants in the mixture. Atrayee et al. study the rheological properties of Viscoelastic Surfactant (VES) based fracturing fluids developed from zwitterionic surfactant Cocamidopropyl betaine (CAPB) mixed with anionic surfactant SDS and sodium oleate (NaOA) respectively [46]. Static and dynamic rheological tests indicate lower CMC and superior viscoelastic properties of the later system (Figure 1).

### Numerical simulation of mixture surfactants

Numerical simulation of mixture surfactants can help us to understand interaction mechanism of different surfactants deeply and comprehensively.

AVai et al. study the characteristics of self-assembly spherical micelles formed by anionic surfactants - polyethylene glycol ethers

in water and sodium chloride solution, respectively [47]. The results show that the addition of salt has a significant effect on the structure and size of the micelles. In pure water, the hydrophobic hydrocarbon chain radicals of the surfactant accumulate in the centre of the micelle; while in sodium chloride solution, the polar group will gather around the micelle crown. Meanwhile, the rearrangement is accompanied by micelle shrinkage, and the spherical micelle radius will be reduced from 2.0nm to 1.8nm.

Wang et al. use molecular dynamics simulation to study the adsorption of cationic surfactant dodecylamine (DDA) and anionic surfactant sodium oleate (NaOL) on the interface [48]. The results show that DDA and NaOL are clearly distributed around the air/water interface, the hydrophobic carbon chain stretches into the air phase, the hydrophilic head group arranges on the air/water interface, and most of the counter ions exist in the interface region near the head group. All heads are hydrated in the aqueous phase, with only a small portion of the carbon chain immersed in the water. Due to the electrostatic interaction between DDA and the NaOL ion-based head group and the hydrophobic interaction between the carbon chains, the mixture displays much more compactness and tightness, thus showing stronger surface activity than the pure DDA and NaOL. When the molar ratio of DDA to NaOL is 1:3, will demonstrate a stronger interaction and a lower interfacial tension, which corresponds well with the results obtained from experimental results.

## Surfactant Combination with Nanoparticles and Polymers

Chemical flooding has been one of the efficient EOR techniques that use polymer (P) and surfactant-polymer (SP) solutions to improve oil/gas recovery by decreasing interfacial tension/surface tension, increasing capillary number, improving mobility ratio, and increasing macroscopic sweep efficiency [49,50]. Nano fluids are of widespread interest in many subsurface engineering applications such as hydrocarbon exploration and production, EOR processes [51-53]. Therefore, Surfactant combination with nanoparticles and polymers is gaining more and more attention.

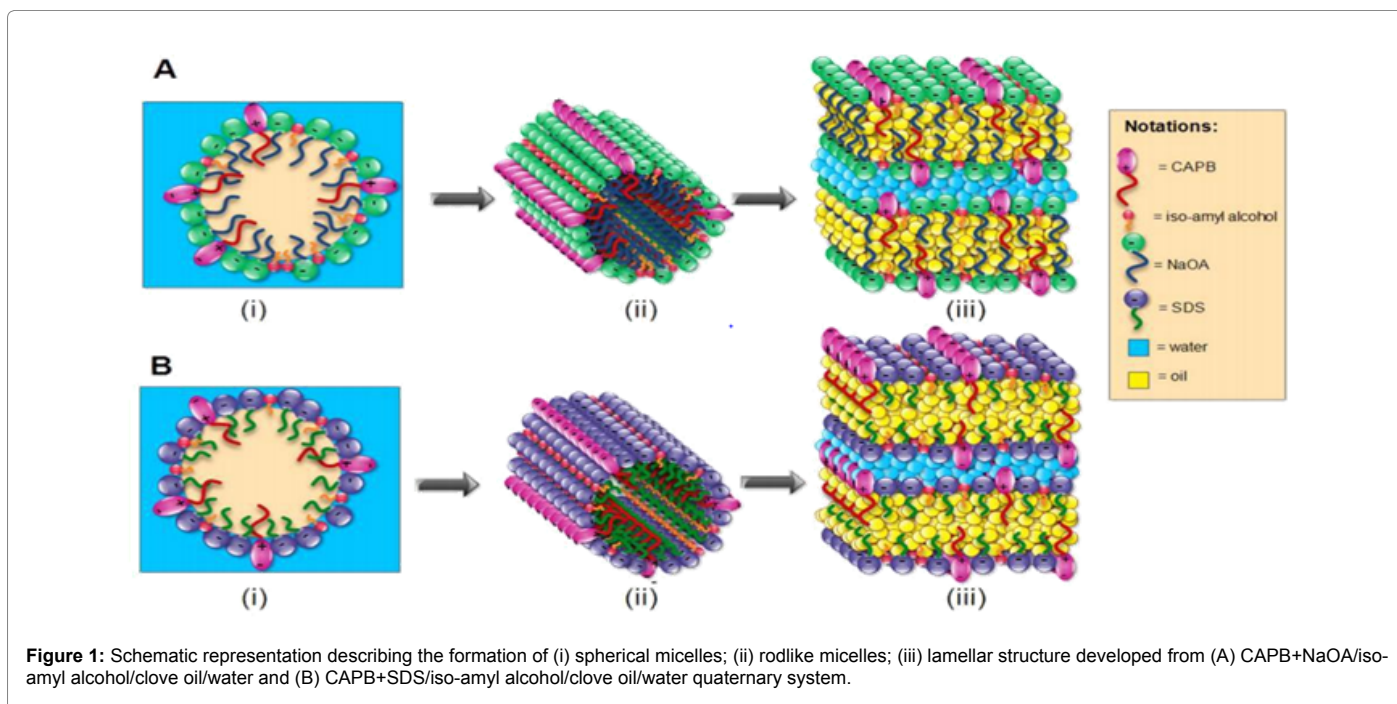
### Surfactant-nanoparticle mixed system

Nanoparticles have a large specific surface area. When added to surfactant solution, nano-emulsions (micro-emulsions) will form and they have better stability due to small-scale effects and Brownian motion. Meanwhile, the nanometer-scale droplets are more likely to enter the tiny voids or cracks of the dense rock, and its large specific surface area can make the nano-droplets fully spread on the rock surface, improve the contact efficiency between the nanoemulsion and the reservoir, and improve the overall application effect of the well fluid [54].

Mohammad et al. compound silica nanoparticles with surfactant surfactant sodium dodecyl benzene sulfonate to prepare NS nanofluids [55]. The interfacial tension between NS nanofluids and crude oil was significantly lower than that of surfactant solution without nanoparticles. It is believed that the electrostatic repulsion between the surfactant and the nanoparticles promotes the diffusion of the surfactant to the interface, while the nanoparticles encapsulated by the surfactant carry the surfactant to the interface due to its Brownian motion, and the surfactant and Surfactant-coated nanoparticles are aligned at the interface to reduce interfacial tension synergistically.

Dong et al. add titanium dioxide nanoparticles directly into water to form nanofluids. As a result, the surface tension of nanofluids





decreased firstly and then increased with the increase of titanium dioxide concentration [56]. It is suggested that when the concentration of titanium dioxide nanoparticles is high enough, the attraction between the particles causes capillary forces, which in turn causes the surface tension to rise. The article also explains the reason for the decrease in surface tension from the energy point of view. It also shows that the reduction of surface tension is also a dynamic process because the adsorption of nanoparticles on the surface is a dynamic process.

Jin et al. study the interaction between surfactant and nanoparticles. cationic surfactant CTAB, anionic surfactant SDS, positively charged titanium dioxide nanoparticles and negatively charged silica dioxide are used in the experiment [57]. The results show that when the surfactant and nanoparticle possess same charge, the electrostatic repulsion will drive more surfactants at the interface, and the two will show synergistic effect, then the surface tension is reduced; While when the surfactant and the nanoparticle possess opposite charge, due to the electrostatic attraction, the nanoparticles will adsorb on the surfactant and agglomerate will form, thereby reducing the effective concentration of the surfactant on the interface, and the surface tension will increased instead.

Luo et al. investigate the effect of clean-up additive compounded by nanoemulsions and surfactants [58]. It is believed that the nanoemulsion has ultra-low surface tension, high solubilisation and small particle size, can effectively enter the pores and fully contact with the solid surface, which can reduce the surface tension and change the surface wetting angle of the solid, then significantly reduce the water absorption of the core, thereby reducing the capillary resistance and effectively improve the fracturing cleanup efficiency.

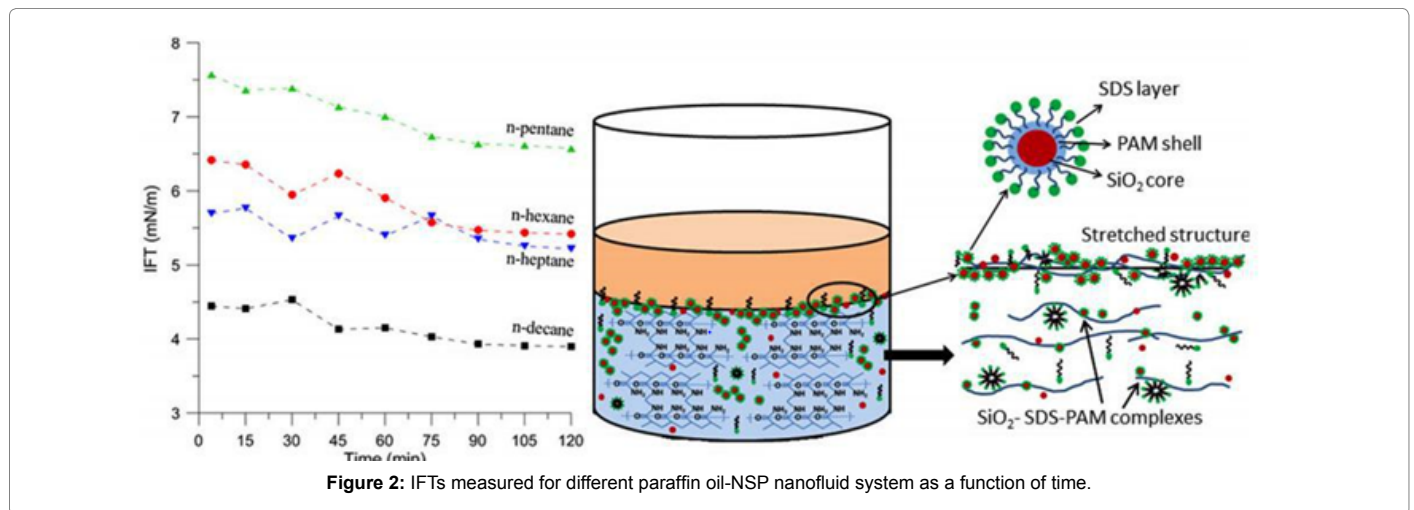
Studies have shown that due to the unique advantages of nanoparticles, the interaction of surfactants and nanoparticles can change the rheological properties and interfacial properties of the system, reducing the surface tension of the system.

### Surfactant-polymer mixed system

When water soluble polymer, surfactant and salt are mixed in aqueous solution, structures known as aggregates may form through polymer-surfactant interaction and can have a drastic effect on the solution's rheology. The structure of these aggregates described as polymer film will form around surfactant micelles [59].

Tushar et al. prepare NP nanofluids by compounding silicon dioxide nanoparticles with polyacrylamide for oilfields, and NSP nanofluids by compounding silicon dioxide nanoparticles, surfactant sodium lauryl sulfate and oilfield polymers polyacrylamide, respectively [60]. The surface tension of nanofluids and the interfacial tension between nanofluids and crude oils are characterized. The results show that the surface tension and oil-water interfacial tension of NP nanofluids and NSP nanofluids are significantly lower than those of traditional oil extraction auxiliaries agent. Furthermore, the surface tension and oil-water interfacial tension of NSP nanofluid are lower than that of NP nanofluid. It is believed that the presence of the surfactant improves the wettability of the nanoparticles, increases the amount of adsorption on the surface, and simultaneously forms the silica nanoparticles-polyacrylamide in the bulk phase due to the Brownian motion of the nanoparticles. The surfactant agglomerates are carried to the surface and arrange directionally on the surface, reducing the surface tension of the system (Figure 2).

Molecular dynamics can be used not only for the simulation of surfactant systems, but also for the study the interaction of surfactant-polymer system to obtain detailed information on the kinetic and structural properties that are not possible in the experiment. Wang et al. use molecular dynamics simulation to study the interaction of cationic surfactant CTAB and the negatively charged polymer sodium polyacrylate (NaPAA) at the air/water interface [61]. The results show that when the surfactant concentration is low, a surfactant-polymer monolayer is formed at the interface, and a multi-molecular layer is formed as the concentration of the surfactant increases. Due to the



electrostatic interaction between the negatively charged carboxylate in acrylic acid and the positively charged head group in the surfactant, the oxygen atom of the carboxylate and the hydrogen atom in the water can form a hydrogen bond, which promotes the binding of the surfactant to the polymer chain.

As the concentration of the surfactant increases, due to the hydrophobic interaction between the hydrophobic tails of the surfactant, the layer-layer arrangement is promoted to form a multilayer adsorption structure. Simulation studies have also shown that the polymer-surfactant complex has a dynamic process of ion exchange, consistent with experimental results.

Studies have shown that due to the interaction of surfactants and polymers, the rheological properties and interfacial properties of the system can be improved.

## Other Applications of Surfactants in Fracturing Process

### Wettability alteration

In gas condensate reservoirs, well productivity reduction resulting from condensate blockage has been one of the most important factors influencing gas and condensate production rates, which is sharper for the low-permeability reservoirs [62]. Many investigators have proposed several methods such as gas recycling, hydraulic fracturing and solvent injection, that is to say using surfactants to mobilize the condensate in the region near wellbore, to restore gas and condensate production rates when condensate blockage occurred [63-65].

In the past few years, many researchers have investigated on surfactants to reduce the surface tension and remove the condensate banking [66,67]. However, these chemicals do not alter the wettability of the rock surfaces from liquid to gas. In recent years, wettability alteration, as a new method, has become more attractive for researchers in industry. Most of the gas condensate reservoirs rocks are naturally liquid-wetting and altering the wettability of the reservoir rock from strongly liquid wetness to preferential gas wetness or intermediate-wetting can increase the mobility of condensate and the relative permeability to gas [68]. Then several researchers develop fluorosurfactants and modify the wettability of rocks from liquid to gas [69-71]. A direct relationship with the increase of roughness and heterogeneity of the surface by means of adsorption of the fluorinated surfactant over the rock surface is discussed. They conclude that the surfactant

adsorption leads to a reduction of the surface free energy due to the formation of a fluorine atoms network [72,73]. Wang et al. investigate the influence of wettability alteration on the displacement efficiency of gas-condensate by conducting contact angle measurement and gas flood tests [74]. Results show that core wettability can be altered from liquid-wetting to intermediate gas-wetting or preferential gas-wetting by the fluorosurfactant at a very low concentration because the core surface free energy sharply decreased after the treatment of surfactants. And then, oil displacement efficiency, relative permeability, and gas flow in gas-oil systems can be effectively improved by fluorosurfactant treatment due to alteration of wettability. Hassanajili et al. synthesize a fluorinated polymer surfactant to induce gas wetness on reservoir rocks, they used static contact angle (CA) tests on thin sections of rock samples; and water/n-decane-air imbibition tests in core scale to demonstrate the applicability of synthesized chemical. Fahimpour et al. investigate the effect of anionic and nonionic surfactant on the wettability alteration of carbonate respectively, results demonstrate that on positively charged carbonate surfaces, the anionic chemicals are sufficiently effective to repel the liquid phase, whereas the nonionic chemicals show an excellent stability in brine media [75,76]. Then a new approach of combining anionic and nonionic chemical agents is proposed.

However, Because of the amphiphilic character of surfactant, they tend to form micelle at certain concentration, which could not penetrate all pores, especially in the tight reservoirs [77]. Therefore, it is urgent to develop chemicals with appropriate size which can diffuse into deeper pore throats in low permeability rocks to alter their surfaces from liquid-wet to gas-wet state.

Nanotechnology has recently emerged as an attractive topic of research in the oil and gas industry due to its exceptional characteristics that allow nanoparticles to travel smoothly through porous media without additional risks of pore blockage due to their small size, which can be used to avoid formation damage. At the nano-scale, exceptional properties can be obtained, such as a high surface-area-to-volume ratio and dispersibility, in addition to high thermal, chemical stability and dispersibility [78]. Hence, the use of nanofluids based on the interaction of fluorosurfactants and nanoparticles with the appropriate particle size could be a promising alternative for enhancing wettability alteration to gas-wet systems.

Jin et al. investigate the influence of gas-wetting alterations on

cores using silicon dioxide nanoparticles modified with fluorocarbon surfactant [79-80]. The results showed that the wettability of the core could be altered to intermediate gas-wetting or super gas-wetting after nanofluid treatment. They conclude that the dehydration-condensation reactions of silicon dioxide nanoparticles and fluorocarbon surfactant forms a compact layer which can remarkably alter the wettability from water-wet to gas-wet (Figure 3).

Maribel et al. develop a nanofluid based on the interaction between an anionic surfactant Silnyl<sup>®</sup>FSJ (SY) and silicon dioxide nanoparticles to alter the reservoir wettability from a liquid-wet state to gas-wettability [81]. The results of different concentration of nanoparticles and surfactant are supported by contact angle and imbibition tests on oil-wet and water-wet sandstone samples for the wettability alteration, then the optimum concentration is achieved and performed under tight gas-condensate reservoir temperature and pressure conditions. The result indicate that the synthesized nanofluid can alter the wettability of the system from a strongly liquid-wet to a gas-wet condition, then the formation damage caused by the condensate banking can be reduced, the production of oil and gas can be considerably improved.

At present, there is only few research concerning the wettability alteration resulted from interaction of nanoparticle and surfactant. Nanofluid combining the benefit of nanoparticle and surfactant may be an effective way to change the wettability from water-wet to gas-wet, which can be a research hotspot in the near future.

### Drag reduction

In the development of shale gas reservoirs, the addition of surfactants can reduce the viscosity and vortex formation in the shear flow of the wellbore, thus reducing the flow resistance of fracturing fluid [82]. At present, the drag reducer added in the fracturing fluid is mainly linear vegetable gum and poly acrylamide, whose shear resistance is poor at higher shear rate, and residue content is high, which may cause serious formation damage [83]. Surfactant is a new type of drag reducer different from polymer, the appearance and disappearance of drag reduction is reversible, and shear degradation does not occur, so it has stable drag reduction effect, especially for large-scale and high shear rate fracturing, as well as the recirculation of fracturing fluid [84]. Studies have shown that when the surfactant drag reducing solution reaches the critical micelle concentration (CMC), spherical micelles are first formed in the solution; with the concentration further increased to the transition concentration (CMCII), the spherical micelles in the solution will transform into rod micelles. When the fluid flows, the rod micelles gradually assemble into a shear-inducing structure and a spatial network structure under shear, then the viscosity of the solution

increases and becomes a viscoelastic fluid [85]. Yu et al. believe that the addition of surfactants in the turbulent flow has a dual role: on one hand, the introduction of viscoelastic shear stress can increase the friction, on the other hand, the suppression of the turbulent structure perpendicular to the wall resulting reduction of turbulent shear stress can reduce the friction [86]. Since the latter is stronger than the former, the surfactant exhibits drag reducing effect when added.

A single surfactant has a limited temperature range and needs large amount in the drag reduction process, which greatly limits its use for drag reduction. Adding surfactants of different alkyl chains together is an effective method to expand the effective temperature range and reduce the amount of surfactants, because it is more favourable to form a stable linear micelle network structure after mixing [44]. The addition of counter ions and salts to the surfactant solution can promote the formation of rod micelles. Tuan et al. have shown that after adding a certain proportion of salt to the surfactant solution, a stable micelle structure can form and intertwine each other [87]. Subsequently, a network structure appear, which exhibit significant viscoelasticity, known as worm-like micelles. Peng et al. add the counter ion salicylate sodium to dodecyltrimethylammonium (CTAB), which is believed to increase the size of the rod micelles and the space size between them [88]. The electrostatic energy between the micelles is reduced, so that the electrostatic stability of entire rod shape micelles in the aqueous solution is enhanced. On the other hand, another part of the non-rod micelles formed for the reason that the hydrophilic groups can not completely cover the hydrophobic groups can be converted to rod micelles, reducing the CMCII value of the surfactant any further.

Cationic surfactants are widely used for drag reduction for the wide effective temperature range. Cationic surfactants CTAC have excellent light, heat and shear stability, has been one of the currently attractive surfactants. Hadri et al. study the effects of temperature and concentration on the drag reduction performance of low concentration cationic surfactant CTAC and counter ion sodium salicylate (NaSl) during turbulent drag reduction [89]. The results showed that when the CTAC concentration is greater than 50ppm and the average temperature is 20°C, the drag reduction rate reaches the maximum. As the temperature increases, to achieve the same drag reduction effect, the CATC concentration needs to be increased. In addition, the zwitterionic surfactant has the characteristics of low toxicity and high biodegradability, which can be combined with an anionic surfactant to obtain a compound reducer with excellent comprehensive performance, as well as the compound of nonionic surfactant and the cationic surfactant. It is also reported that a kind of a drag reducer composed of twelve alkyl betaine and sodium twelve alkyl sulfate and another kind of

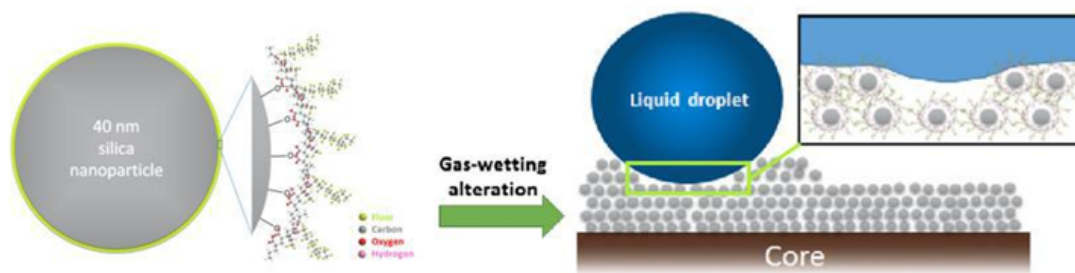


Figure 3: Sketch of surface modification of a silica nanoparticle and its gas-wetting alteration application.



drag reducer composed of rapeseed oil oleic acid ethanolamide, lauryl alcohol and sixteen alkyl three methyl salicylic acid ammonium salt both show strong drag reduction performance [90].

When drag reduction occurs for the surfactant solution, the turbulent structure inside the fluid will be inhibited by the induced structure formed by the surfactant, thereby changing the turbulent structure, which provides possible conditions for other drag reduction methods that acquiring drag reduction by influencing turbulent structures, and also provides guidance for combining surfactant drag reduction with other suitable drag reduction methods. Therefore, the study of surfactants combined with other drag reduction methods to realize efficient turbulence drag reduction is also a hot topic in the near future [91].

A nanoparticle-associated surfactant micelle fracturing fluid has been developed recently, which can associate linear micelles to form a three-dimensional network structure. Nanoparticles at very low concentration can greatly improve low shear velocity viscosity, swelling time and elastic storage modulus. It is believed that the surface of the nanoparticle can interact with the end cap of the linear micelle, functioning as a joint bond, then the micelles will be coupled into a micelle network. The nanoparticle can replace the end cap and join the nanoparticle and the worm-like micelle [92].

Maxey et al. study the effect of nanoparticles on improving the rheological properties and fluid loss control properties of surfactant fluids, indicating that surfactant fluids without nanoparticles exhibit significant viscosity, while the surfactant fluids will show significant elasticity as soon as the nanoparticles were added [93]. It is suggested that the addition of nanoparticles increases fluid shear viscosity, improves fluid loss control, and improves shear stability due to the formation of a viscous network structure, exhibiting similar properties to crosslinked polymers. In general, the addition of nonionic surfactants to nanofluids is the best choice because of the negative effects of nanosuspension settling and nanoparticle settling is least.

Matras et al. study the effect of the interaction of nonionic surfactant cocoamidopropyl betaine with high molecular weight polymer polyoxyethylene and anionic polyacrylamide on the drag reduction effect through the parameters like surface tension, viscosity, and electrical conductivity, respectively [94]. The results show that due to the hydrophobic interaction and electrostatic interaction between the high molecular polymer and the surfactant, a polymer film is formed around the surfactant micelle to generate a polymer-surfactant agglomerate, which changes the flow characteristic of the original system. The novel suspension shows higher drag reduction rate, larger range of drag reduction Reynolds number and effective temperature range, stronger shear resistance, and partial recovery after shear failure.

Liu et al. test the mixed aqueous solutions of cationic surfactant CTAB, non-ionic molecular polymer polyacrylamide and NaSal as the counter-ion salt to verify the speculation about their intensification possibilities of drag reduction performance under different conditions [14]. The research show that surfactant molecules form micelles round polymer chains, then a kind of reinforced concrete structures will form (as shown in Figure 4) with the increase of Reynolds number, but exceed which the reinforced concrete structures will be destroyed, the formed reinforced concrete structures were more complex and more effective in restrain vortices, leading to the intensification of drag reduction compared to pure surfactant solutions, the experiment also prove that temperatures were more influential than concentrations.

MalcherT et al. Investigated the drag reduction mechanism and properties of nonionic polyoxyethylene(PEO) and cationic surfactant CTAB agglomerates by computational fluid dynamics Numerical simulation. The results show that the complex structure of CTAB-PEO agglomerates of neutral polymer and surfactant are non-covalently bonded in aqueous solution, with a viscosity higher than that of single surfactant or polymer. In a certain range of Reynolds number, agglomerates will elongate, stretch and transfer internal stress when subjected to shear in the flow, thereby reducing the resistance.

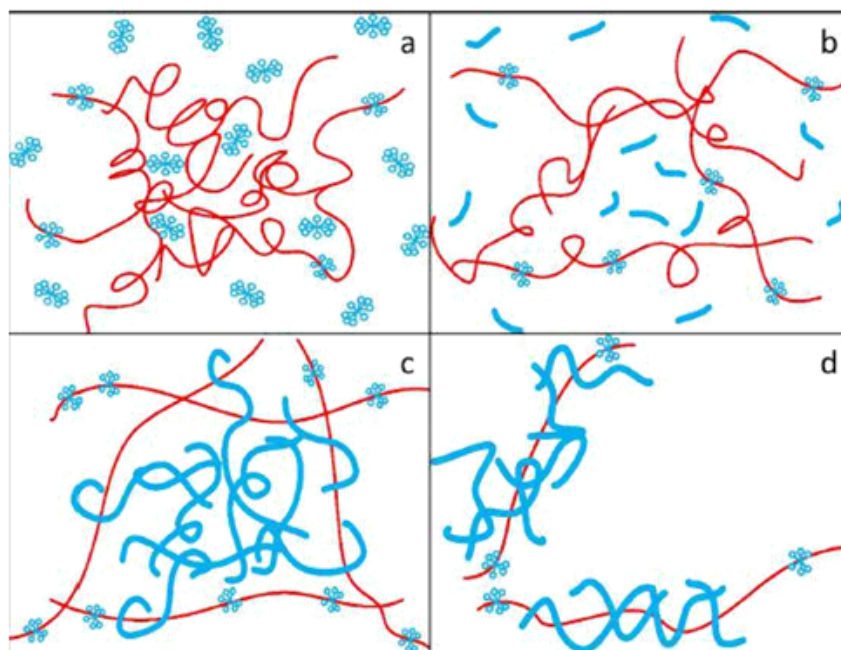


Figure 4: The formation and destruction of reinforced concrete structure by polymer-surfactant.

Studies have shown that surfactants can present superior drag reduction effect with nanoparticles and polymers, synergistically. It can retain the advantages of a variety of original single materials, make up for its shortcomings, improve drag reduction efficiency and reduce drag stability, which will be a hot spot in the future research of drag reducers (Figure 4).

## Summary and Prospect

The up-to-date progress of the application of surfactants in fracturing fluids for enhancing oil/gas production are summarized comprehensively from the aspects of single surfactant system, mixed surfactant system, surfactant combination with nanoparticles and polymers, mainly concentrating on the effect of surface tension reduction, wettability alteration and drag reduction from the point of experimental research and numerical simulation. Based on the research on the surfactants summarized above, surfactants, as one of the most important clean-up additive in fracturing, have multiple functions including reducing surface tension, altering wettability, reducing flow friction and so on. However, the effect of single surfactant is poor, which can be improved by the compounding system. Nanoparticles have the advantage of large specific surface area, small particle size, and obvious Brownian motion effect, which can show significant effects when compounded with surfactants. Polymer-surfactant aggregates can have a drastic effect on the solution's rheology. Therefore, the study on the mechanism and interaction of surfactants, nanoparticles, and polymers in Nano fluids will be the hotspot of research on fracturing fluids in the future.

Hydraulic fracturing consumes a huge amount of water. Faced with the current dual pressure of development cost and environmental protection, it requires 100% reuse of fracturing fluid. The exploitation of "green"clean-up additive with high flow back rate, excellent resistance, and low damage to formation under high salinity to meet the requirements of on-site fracturing construction will be the focus of future research and will also have broad application prospects.

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