Colloids and Surfaces A 550 (2018) 176–185

Contents lists available at ScienceDirect

Colloids and Surfaces A

journal homepage: www.elsevier.com/locate/colsurfa

Synergy of surface-treated nanoparticle and anionic-nonionic surfactant on stabilization of natural gas foams

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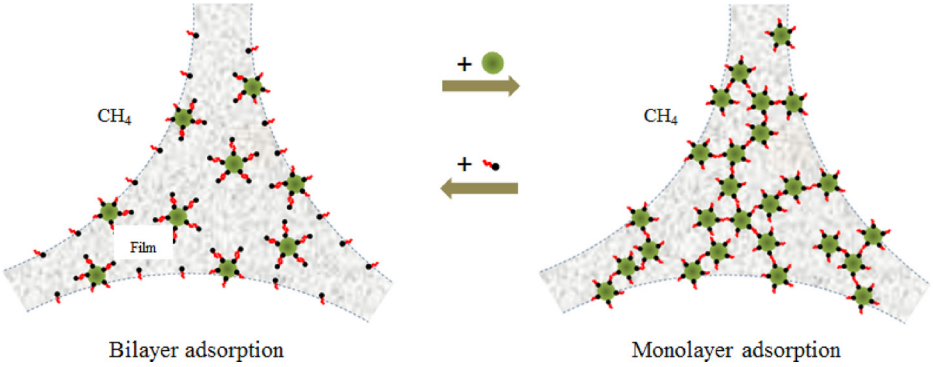
GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Positively charged nanoparticle
Anionic-nonionic surfactant
Natural gas foams
Stability
Surface adsorption
Synergy

ABSTRACT

Positively charged surface-treated nanoparticle (S-AK) and anionic-nonionic surfactant sodium fatty alcohol polyoxyethylene ether sulfate (AES) mixture has been used to obtain natural gas foams. It was found that synergy occurs between S-AK and AES in generating stable natural gas foams due to the foam stability of the S-AK/AES mixture being better than that of either single component. It is important to control the contents of two species since there are optimum concentrations for S-AK and AES on stabilizing Natural gas foams. High salinity ($\geq$50,000 mg L^{-1}) is conducive to the stability of S-AK/AES natural gas foams at high temperature (50 °C). In addition to the electrostatic interaction, EO groups significantly promote the attraction between S-AK and AES. The stability of S-AK/AES natural gas foams may relate to two aspects: one is that AES-adsorbed nanoparticles migrate to the gas/liquid interface and enhance the interfacial dilatational elasticity, and the other is that a rigid skeleton structure forms in the natural gas foam films by the flocs consisting of S-AK and AES. Natural gas foams are most stable when the monolayer adsorption of AES molecules on the S-AK surface occurs in the dispersion.

1. Introduction

It is well known that gas injection is one of the popular enhanced oil recovery (EOR) methods [1]. However, immiscible gas injections, such as air, CO_2 or N_2 injection, for EOR rarely exhibit good sweep efficiency because of high gas mobility and reservoir heterogeneity [2]. Foam

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https://doi.org/10.1016/j.colsurfa.2018.04.046
Received 4 March 2018; Received in revised form 18 April 2018; Accepted 19 April 2018
Available online 23 April 2018

flooding is an EOR technology to overcome the problems of immiscible gas injections by dispersing bubbles in a continuous aqueous phase thus to increase the apparent viscosity of the gas phase and increase the fluid sweep coefficient in reservoirs [3]. Due to the large gas/liquid interface area and high surface free energy, foam is actually a thermodynamically unstable system and always ruptures in a short time [4]. Therefore, it is very necessary to improve foam stability for its application.

At present, much research on foaming agents is mainly focused on the synthesis or compounding of surfactants [5]. Though these foaming agents produce enough foam volume, the disadvantage is the poor stability of the foams. Polymers, such as polyacrylamide and xanthan gum, play a role in enhancing the stability of the foams, but the foam volume is low [6]. Poor temperature tolerance or salt tolerance of the polymers also makes the foams easy to break down, which is not conducive to the application of foams in high temperature or high salt environments [7].

Solid particles, such as oxides, clay particles and $CaCO_3$ particles, have already been confirmed to improve the stability of the foams, the so-called Pickering foams [8,9]. The stability of Pickering foams is affected by a variety of factors, among which particle size is a major influencing factor that has been studied in depth [10–12]. Tang et al. [13] found that fine silica particles could improve the stability of the sodium dodecyl sulfate (SDS) foams by investigating particles of a wide particle size (20–700 nm in diameter). Kam et al. [14] thought that small size particles had high adsorption energy and produced films with a large rigidity to inhibit the Ostwald ripening of the foam. Wasan et al. [15] believed that when large particles were added to a mono-dispersed small particle environment, the large particles always arranged near the film wall due to osmotic dissipation forces, and water flowed around the large particles, resulting in localized bending and thinning of the film, such that foam stability decreased.

Currently, nanoparticles stabilizing the foam are receiving more and more attention. SiO_2 nanoparticles with diameters between several and several tens of nanometers are the most widely used. Generally, bare SiO_2 nanoparticles are difficult to be used to stabilize the foams due to their hydrophilicity. Dickinson et al. [16,17] reported that partially hydrophobic SiO_2 nanoparticles could stabilize CO_2 foams. Binks et al. [18] changed the hydrophobicity of the SiO_2 nanoparticles containing SiOH groups by reacting with silanizing agents and found that the nanoparticles containing 32% SiOH on the surface had the best foaming properties. Zhang et al. [19] prepared stable CO_2 foams by partially hydrophobic SiO_2 nanoparticles and sulfosuccinate and thought these CO_2 foams were the most stable when the gas/liquid ratio was 5:1.

Some researchers believed that the main reason for particles stabilizing foams was that the particles increased the effective viscosity of the continuous phase liquid [20,21]. Fyrillas et al. [22] theoretically illustrated that the formation of gel in the liquid film increased the storage modulus of the liquid film and inhibited the Ostwald ripening caused by the gas diffusion. Most researchers thought that the stabilization of the foams by the particles was mainly attributed to the adsorption of the particles at the gas/liquid interface and that the particles acted as an elastic separator to separate the bubbles [23,24]. Irreversibly adsorbed particles formed a solid shell to stabilize the foams, as shown by Kam et al. [14]. Binks also came to a similar conclusion that the formation and stability of the foams depended on the balance between the adsorption of hydrophobic particles on the gas/liquid interface and the aggregation in the bulk [25]. The foamability of the hydrophilic particles is low, and the stable foams formed only when partially hydrophobic particles had a strong tendency to aggregate in bulk. Dickinson et al. [26] observed that the aggregated particle layers of the bubble surface were connected to the three-dimensional weak-gel particle layers formed in the bulk by laser confocal microscopy. A few studies also argued that non-adsorbed particles had a stabilizing effect on the foams. As long as the size of the foam film was within a certain range and the particles were just smaller than the initial thickness of the liquid film, the particles could be drilled into the liquid film and were difficult to discharge [15].

Although there are many studies on nanoparticle-stabilized foams, the nanoparticles are mostly hydrophobic surface-modified nanoparticles. Studies on other types of nanoparticles used to stabilize the foams have been rarely reported. Furthermore, most research has mainly focused on the nanoparticle-stabilized foams formed by air, CO_2 and N_2. Natural gas is abundant in some oil reservoirs, facilitating their use in those places. Hydrocarbon components of natural gas and water-based foaming agents are more prone to foam, and the oil viscosity reduces when the natural gas dissolves in the oil. The above advantages are conducive to the development and application of natural gas foams in tertiary oil recovery. However, limited studies of natural gas foams have been reported to date. Therefore, in this study, it is tried to generate natural gas foams by positively charged surface-treated nanoparticles and clarify the mechanism between this surface-treated nanoparticle and an anionic-nonionic surfactant in stabilizing the natural gas foams. The study is expected to provide a direction for the design or synthesis of nanomaterials with better natural gas foam stability, which is of great theoretical and practical value for the application of natural gas foam technology in oil exploitation.

2. Experimental

2.1. Materials

Sodium fatty alcohol polyoxyethylene ether sulfate (AES, CH_3-$(CH_2)_{11}$-O-$(CH_2CH_2O)_3$-SO_3Na), the anionic-nonionic surfactant, was purchased from Chengdu Aike Chemical Technology Co., Ltd., China, with an active content of 70%. The critical micelle concentration (*cmc*) of AES is 74 mg L^{-1} and the surface tension at *cmc* (γ_{cmc}) is 31 mN m^{-1} in pure water at 25 °C. Sodium dodecyl sulfate (SDS, $CH_3(CH_2)_{11}OSO_3Na$), the anionic surfactant, was kindly provided by Stepan Co., USA, with an active content of 97%. The *cmc* of SDS is 2365 mg L^{-1} and theγ_{cmc} is 25 mN m^{-1} in pure water at 25 °C. Silica nanoparticles (S-AK) with an average particle size of 10–15 nm were received as a 17.6 wt% aqueous dispersion at a pH of 4.5 from Nissan Chemical Industries Co., Ltd., Japan. S-AK nanoparticles were positive charge surface treated with Al_2O_3 and a specific surface area of 230 m^2/g and a specific gravity of 1.141. NaCl and $CaCl_2$, purchased from the MP Biomedicals, LLC., USA, were all of reagent grade. Methane (CH_4, 99.97% purity) was used to represent natural gas and supplied by Praxair Technology Inc., USA. All samples were used without further purification.

2.2. Preparation of the foaming agents

The concentrated S-AK dispersion was first subjected to ultrasonic oscillation for half an hour to obtain uniformly dispersed nanoparticles. The concentrated AES solution was prepared by dissolving an appropriate amount of original sample in pure water. Then, a series of lower concentrations was obtained by diluting the concentrated solution. The foaming agents were prepared by adding S-AK dispersion to AES solution and gently shaking without foams generation.

2.3. Forming test and stability determination of natural gas foams

Based on the Ross-Miles method [27], an improved method called Jet method was designed to generate natural gas foams. The Jet method is a convenient way to dertermine the foam stability in a confined space, which is more suitable for the gases other than air. The main procedure of the Jet method is shown in Fig. 1:

(1) Put a graduated cylinder in a temperature-controlled water bath and fill it with water. Then replace water with methane from the methane source by the drainage method. The graduated cylinder