



Development, formation mechanism and performance evaluation of a reusable viscoelastic surfactant fracturing fluid



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ABSTRACT

Through extensive experiments, a reusable viscoelastic surfactant (rVES) fracturing fluid using a self-designed and synthesized surfactant was developed. Laboratory performance evaluation results showed that various performances of rVES fracturing fluid are excellent. The most attractive aspect is the gelling and gel breaking of rVES fracturing fluid was achieved by altering the pH value. The recycled fracturing fluid still maintains good performances through three times cycles, which represents that it has good prospects for re-use of flowback waters. In addition, large-scale coarse-grained molecular dynamics (CGMD) simulations results clearly revealed the formation mechanism of rVES fracturing fluid at the molecular level.

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Introduction

The use of hydraulic fracturing technology has been a well-established practice for enhancing productivity during the development of low-permeability reservoirs for more than 60 years [1]. The major functions of the fracturing fluid are creating fractures in reservoir and transporting proppant to the fracture to maintain a high conductivity of formation [2]. Polymer-based fluids, including guar or polyacrylamide, have constituted the majority of polymers in fracturing fluids in this decade. Nevertheless, the greatest shortcoming of polymer-based fluids is that they can cause formation damage, which is a serious problem with detrimental effects on production enhancement [3].

Viscoelastic surfactant (VES) fracturing fluids have attracted considerable scientific and practical attention and have been used in a variety of applications, including hydraulic fracturing, matrix acidizing, gravel packing, drilling, and coiled tubing cleaning [4]. Compared with conventional polymer fracturing fluid, VES fracturing fluid has many advantages, such as few components, easy preparation, good prop-carrying capacity, low treatment friction, no residue, low damage to formation, and automatic gel-breaking [5]. The viscoelasticity of VES fracturing fluids is due to

the formation of entangled wormlike micelles [6]. In recent years, smart wormlike micelles, whose structure can be manipulated by external stimuli [7], have also progressed greatly. The initial VES fracturing fluids took advantage of two different reservoir conditions (external conditions) to break the VES fracturing fluid after the treatment flows back upon the completion of production [8]. The first condition is a change in the brine concentration. VES fracturing fluids are found to be stable over a particular concentration of salts; by diluting this concentration with produced fluids, the gel can be broken. The second condition that can break VES fracturing fluids is their contact with reservoir hydrocarbons. When hydrophobic substances such as oil or gas are dissolved in the hydrocarbon core of the micelle, the structure becomes swollen and then breaks into smaller spherical micelles. These two conditions may not be met in all cases. For example, when reservoirs do not produce hydrocarbons, the VES fracturing fluids does not break efficiently. Hence, an internal VES-breakers technology was developed to address the above-mentioned issues. Two breaking mechanisms that have been utilized by internal VES-breakers technology are presented [9]. The first mechanism is adding compounds that can generate low molecular weight alcohols and thereby degrade the wormlike micelles to non-viscous spherical micelle. The second mechanism is adding oxidant breaker to destroy the structure of surfactant molecules so they can no longer act as a surfactant. The two treatments are nonreversible, and viscosity cannot be restored to the produced fluids for the recycling of the fracturing fluid flowback.

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In the present work, we developed a reusable viscoelastic surfactant (rVES) fracturing fluid. The performances of rVES fracturing fluid resistance to temperature and shearing, proppant suspending ability, salt tolerance properties, viscoelasticity and reservoir protection were evaluated through extensive experiments. Furthermore, two gel breaking methods were compared by investigating the effects of hydrocarbons, brine and pH on the viscosities of rVES fracturing fluid. In addition, the performances of rVES fracturing fluid resistance to high temperature and shearing after several repeated cycles was characterized to evaluate the prospects of re-using water and the gelling agents within the flow-back fluid. Finally, CGMD simulations were employed to investigate the formation mechanism of the self-assemblies in rVES fracturing fluid.

Experimental

Materials

2-Docos-13-enoylamino-pentanedioic acid (DEPA) was synthesized as follows. Methylene dichloride, oxalyl chloride, *N,N*-dimethylformamide (DMF), ethyl acetate and potassium hydroxide (KOH), sodium chloride (NaCl), ammonium persulfate (APS), calcium chloride (CaCl₂), magnesium chloride (MgCl₂), all of analytical grade, were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Erucic acid (90%) and citric acid (>99.5%) were purchased from J&K Chemical Ltd. (Beijing, China). Concentrated hydrochloric acid (HCl) was obtained from Hwei Chemical Co. (Shanghai, China). DL-glutamic acid (98%) and Stearyl dimethylbenzylammonium chloride (1827) were purchased from Macklin Biochemical Co., Ltd. (Shanghai, China). Deionized water was used in all measurements.

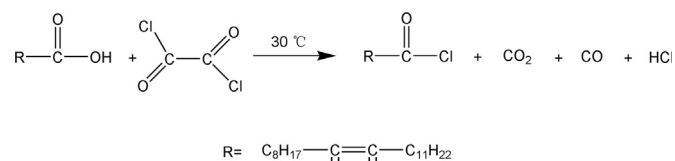
Synthesis of DEPA

Synthesis of erucacyl chloride

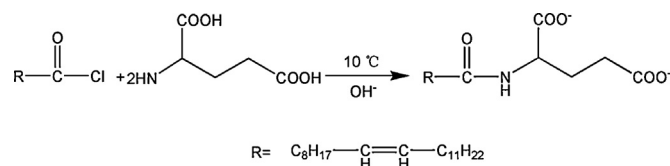
Methylene dichloride (70 mL) and erucic acid (7.51 g, 0.02 mol) were added into a single-necked flask equipped with a drying tube. Oxalyl chloride (3.81 g, 0.03 mol) was added dropwise into the flask and stirred at 30 °C for 1.5 h. Methylene dichloride and residual oxalyl chloride were removed by vacuum distillation. The reaction process is outlined in Scheme 1.

Synthesis of DEPA

DL-Glutamic acid (4.41 g, 0.03 mol) was dissolved in DMF (15 mL). Next, erucacyl chloride was added dropwise with vigorous stirring at 10 °C. During the reaction process, the pH was kept within a range of 9–11 with KOH. Next, the mixture was stirred at 70 °C for another 3 h. The solvent was removed by vacuum distillation, and then a mixture of ethyl acetate (150 mL) and deionized water (50 mL) was added to the residues. Subsequently, the pH of the solution was adjusted to 1–2 by adding concentrated hydrochloric acid. Ethyl acetate and deionized water were removed by vacuum distillation, leaving a white solid. The white solid powder was recrystallized from ethyl acetate three times. The reaction procedure is outlined in Scheme 2. Mass spectrometer measurements were performed using a quadrupole



Scheme 1. Synthesis pathway for erucacyl chloride.



Scheme 2. Synthesis pathway for DEPA.

time-of-flight (Q-TOF) Agilent 6510 mass spectrometer (Palo Alto, US) in the positive-ion mode. The data acquisition was under the control of Mass Hunter workstation software. ¹H nuclear magnetic resonance (¹H NMR) spectroscopy analysis was obtained from a BRUKER AVANCE III HD 400 spectrometer (Bruker, France) operated at 400 MHz. The mass spectrogram and ¹H NMR spectroscopy results are shown in Figs. 1 and 2.

Preparation of fracturing fluid

First, 3 wt% DEPA and 3 wt% 1827 were dissolved completely in distilled water individually. Next, KOH and HCl were used to regulate the pH values of samples. The concentrations of guar gum and polymer in the fracturing fluid were 0.3 wt% and 0.5 wt%, respectively. The samples were placed in a thermostat water bath at 25 °C for 1 week to remove any air bubbles.

Rheology measurement

Rheological properties were determined using an Anton Paar MCR 302 rheometer (Graz, Austria) equipped with a CC39/HA cylindrical rotor (SN: 40522). All samples were equilibrated for at least 15 min before the tests. A solvent trap was used to minimize sample evaporation.

Formation damage evaluation

The core permeability was tested to evaluate the formation damage of the rVES fracturing fluid. A technique for measuring the pore volume of the core sample involves dividing the difference in its weight when dry and when saturated with standard brine by the density of standard brine. The overburden pressure of the core holder unit was fixed at 15 MPa and placed in the 90 °C constant temperature bath pot.

To achieve the applicability of Darcy's law, a precision piston pump was employed to control the injection rate, *Q*, which was set to 1 mL/min. The pressure difference (ΔP) of core was recorded by pressure sensors that were connected to a computer. The permeability *K* was calculated by Darcy's law after the pressure difference stabilization:

$$K = \frac{Q\mu L}{A\Delta P} \quad (1)$$

where *A* is the cross-sectional area of the core, μ is the viscosity of the fluid and *L* is the length of the core. The permeability damage rate of the core (Φ) can be calculated by the following formula:

$$\Phi = \frac{K_0 - K}{K_0} \quad (2)$$

where *K* is the permeability of the core measured by gel breaking fluid and *K*₀ is the initial permeability measured by standard brine.

Molecular dynamics simulation

CGMD simulation was employed to investigate the formation mechanism of the self-assemblies in the rVES fracturing fluid. The equilibrium system size is *L* = 20 nm. The molecule numbers of

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