



Research paper

Effect of polyampholyte-bentonite interactions on the properties of saltwater mud

Ling Lin^{a,*}, Pingya Luo^b^a School of Chemistry and Chemical Engineering, Southwest Petroleum University, Chengdu, Sichuan 610500, People's Republic of China^b State Key Laboratory of Oil and Gas Reservoir Geology and Exploitation, Southwest Petroleum University, Chengdu, Sichuan 610500, People's Republic of China

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ABSTRACT

Hydrolyzed poly(acrylamide/dimethyl diallyl ammonium chloride) was synthesized via free-radical copolymerization followed by hydrolyzation. The synthesized polyampholyte was characterized by Fourier transform-IR and ¹H NMR measurements. An intensive investigation was carried out to identify the effect of salt, temperature, pH, polymer composition, and intrinsic viscosity on the adsorption of the polyampholyte samples on bentonite, the bentonite particle size distribution, and the filtration of saltwater mud. In the mud samples, the polyampholyte interacted with bentonite via adsorption. By introducing electrostatic repulsion and entropic repulsion, the polyampholyte helped maintain a broad bentonite particle size distribution, allowing the bentonite to form a densely packed filter cake after aging treatment. In summary, using a polyampholyte with a high cationic group content, a high intrinsic viscosity, and an anionic group content in a specific range resulted in a strong adsorptive ability on bentonite and allowed ideal control over the clay particle size distribution and mud filtration behavior.

1. Introduction

Water-based drilling fluids (muds) have been extensively used in the oil and gas (O&G) industry for > 150 years (Dai et al., 2011; Smith et al., 1996; Wu et al., 2012; Zhang et al., 1999; Zhang and Yin, 1999). Among the polymeric agents added to water-based muds, filtration additives play an essential role in adjusting the bentonite (Bent) particle size distribution and controlling the fluid loss of muds, especially in harsh conditions such as high temperature, high salinity, and high pressure (Collette et al., 1994; Dai et al., 2011; Liu et al., 2016; Mao et al., 2015; Peng et al., 2010; Smith et al., 1996; Wu et al., 2002; Wu et al., 2012).

Amphoteric polymers, which have both cationic and anionic groups distributed along the same chain, have been frequently used as filtration reducers in water-based drilling fluid because of its active interactions with clay and impressive filtration control at temperatures higher than 180 °C (Bai et al., 2015; Chu et al., 2013; Lin and Luo, 2015; Liu et al., 2016). The interaction between the amphoteric polymers and Bent, mainly in the form of adsorption, depends substantially on the polymer properties such as detailing magnitude of electric charges (both positive and negative), charge distribution along the polymer backbone, and intrinsic viscosity. Anionic groups contribute significantly to the hydration capability of the polymer in aqueous

solution. Furthermore, both cationic group content and intrinsic viscosity play a decisive role in the adsorption of the polymer chains on the negatively charged particles, such as silica in deionized water (Joanny, 1994; Khan et al., 2001; Kudaibergenov, 2008). The adsorptive behavior of polyampholyte should be different from that of polyanion and polycation, as the amphoteric polyacrylamide's adsorption isotherms on bentonite were located in a region between cationic and anionic PAMs under room temperature in the absence of electrolytes (Shaikh et al., 2017).

The adsorption of polyampholyte on charged particles/surfaces is one of the classic research domains in polymer physical chemistry (Dobrynin et al., 1998; 1999; 2001; Joanny, 1994; Khan et al., 2001; Kudaibergenov, 2002; Kudaibergenov and Ciferri, 2007; Musabekov et al., 1998). Previous simulations of the adsorption of polymer chains and its conformations have indicated that the cationic groups occupy most of the adsorptive sites on the planar, negatively charged interlayer surfaces of Bent, while the negatively charged segments extend away from the surface in a pole, fence, or pancake regime (Dobrynin et al., 2004; Kudaibergenov, 2002).

Among the published work concerning polyampholyte adsorption, the most frequently used adsorbents are planar, whereas amphoterically charged adsorbents, such as Bent, have infrequently been used. Increasing the salinity can promote multilayer polyampholyte

* Corresponding author.

E-mail address: cwobolinling@swpu.edu.cn (L. Lin).<https://doi.org/10.1016/j.clay.2018.07.012>

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adsorption rather than monolayer adsorption, resulting in the desorption of chains (Berre et al., 2001). The adjustment of the pH, especially near or far from the isoelectric point (IEP), may lead to transitions between the random coil and granular conformations, affecting the adsorption of the polyampholyte (Song et al., 2010; Tran et al., 2006). Few previous investigations have focused on the adsorption of polyampholyte under harsh conditions such as high temperatures.

Bent particles, if mainly composed of smectite, carry two kinds of electrical charges: a structural negative charge owing to isomorphous substitutions within its structure (Avena and Pauli, 1997). The charge on the interlayer surfaces is usually considered to be permanent and negative, resulting from isomorphous substitutions and independent of pH, while the charge on the edge is pH-dependent, arising from the protonation and deprotonation of surface hydroxyls (Lützenkirchen et al., 2012). Insufficient effort has been made to identify the relationship between polyampholyte adsorption on Bent and the properties of saltwater mud. However, it is important to identify the relationship among the polyampholyte composition, intrinsic viscosity, and adsorption on Bent, and the properties of saltwater mud to determine if the synthesized polyampholyte is an ideal filtration additive for saltwater mud working under high temperature. A related paper investigated the effect of polymer composition, chemical structure of monomeric units in the backbone and charge distribution on rheological and filtration properties of Bent/polymer dispersions under 25 °C and 85 °C, using only anionic polymers (Ahmad et al., 2018). However, few papers were focused on the harsh conditions such as high temperature.

To reveal the influence of the polymer composition and intrinsic viscosity on the interactions between the polyampholyte and Bent, as well as to show the effect of polyampholyte adsorption on the Bent particle size distribution and mud filtration under high temperature up to 180 °C, we have carried out a series of experiments. First, a batch of “acrylamide and dimethyl diallyl ammonium chloride” copolymers with varied anionic group content, cationic group content, and intrinsic viscosity was synthesized. Then these amphoteric polymers were tested to investigate the relationship between their composition, intrinsic viscosity, and the properties of the saltwater mud.

2. Materials and methods

2.1. Materials

Acrylamide (AM), NaHSO₃, ammonium persulfate (APS), MnCl₂·4H₂O, methyl orange, indigo carmine, AgNO₃, K₂CrO₄, sodium formate, sodium acetate trihydrate, cadmium iodide, soluble starch, acetic acid, bromine water (3 wt%), NaOH, Na₂SO₃, Na₂CO₃, NaCl, and ethanol were provided by Chengdu Kelong Chemical Company, China. Dimethyl diallyl ammonium chloride (DADMAC, 60 wt% in water) was provided by Sinopharm Chemical Reagent Company, China. All chemicals above were of analytical reagent grade. Bent for preparing the mud samples was purchased from Zhongfei Xiazijie Bentonite Company, Xinjiang, China. The components in Bent are listed in Table 1, as per China's O&G Industrial Standard SY/T 5163-2010.

The X-ray diffraction (XRD) data of Bent was shown as Fig. 1. XRD test was performed using X'PERT PRO MPD X-ray diffractometer (Netherlands), operated at 40 kV and 150 mA with Cu-Kα radiation ($\lambda = 0.154056$ nm) at a scanning speed of 0.02°/s and the scanning ranges were from 3° to 80°. The cation exchange capacity (CEC) of this Bent was 75.31 cmol·kg⁻¹, as per China Industrial Standard JCT 593-1995. All of the materials listed above were used as received.

2.2. Synthesis and characterization of the polymers

The synthesis of the polymer followed our previously published polymerization and hydrolyzation procedure (Lin and Luo, 2015).

First, 20 g AM and 10–20 g DADMAC were dissolved in 70–93 g

Table 1
Composition of Zhongfei Xiazijie Bent.

Chemical composition (wt%)	SiO ₂	65.13
	Al ₂ O ₃	14.55
	CaO	1.58
	MgO	1.94
	K ₂ O	2.36
	Na ₂ O	5.07
	Fe ₂ O ₃	4.32
	TiO ₂	0.59
	Total	95.54
	Mineralogical composition (wt%)	Quartz (Qtz)
Plagioclase (Pl)		1.84
Smectite (Sm)		69.13
Illite (Ill)		4.71
Kaolinite (Kaol)		2.36
Illite/smectite (Ill/Sm)		2.36

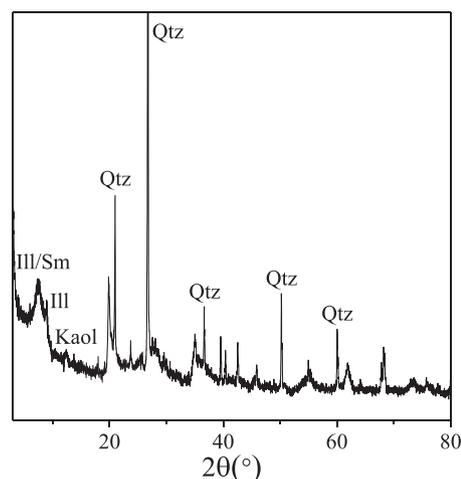


Fig. 1. XRD result of Bent Sample.

deionized water in a four-neck round bottom flask equipped with a thermometer, nitrogen inlet tube, and magnetic rotor. The solution was stirred for 20 min while sparging with nitrogen to remove oxygen, followed by heating to 50 °C in a water bath. Next, a deionized water solution of APS and NaHSO₃ (1:1 M ratio) was added dropwise to the above solution, and the reaction proceeded for 10 h. After the reaction had completed, the poly(acrylamide/dimethyl diallyl ammonium chloride), henceforth PAD, was purified by precipitation in ethanol three times, vacuum filtered, and dried for 24 h at 60 °C under vacuum (10⁻³ Torr).

Subsequently, 10 g PAD and 1.82–5 g NaOH dissolved in 200 g deionized water were transferred to a one-neck round bottom flask equipped with a reflux condensation tube. The flask was heated to 85 °C in a water bath for 24 h. After the reaction between NaOH and amide groups of PAD had completed, the hydrolyzed poly(acrylamide/dimethyl diallyl ammonium chloride), henceforth HPAD, with different content of carboxyl groups was obtained. The hydrolysis degree of HPAD was controlled by using varied amounts of NaOH to react with the amide groups of PAD under high temperatures. The HPAD samples (HPAD-1, HPAD-2, ..., HPAD-9) were purified and dried as for PAD. Please note that the HPAD-10 sample was polymerized and hydrolyzed following the same procedure of other nine samples, except for using “MnCl₂-O₂-NaHSO₃” as the initiator rather than “APS-NaHSO₃”.

The intrinsic viscosity was determined with an Ubbelohde capillary viscometer in NaCl solution (1 mol·L⁻¹) at 30 ± 0.1 °C in accordance with China Standard GB/T 1632-1993. Three replications were conducted for each intrinsic viscosity test, and the variation for t₁ (the flow time for the experimental solution in Ubbelohde capillary viscometer) was < 0.2 s. The cationic group content was measured by titration

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