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Letter

Novel hydrophobic associated polymer based nano-silica composite with core–shell structure for intelligent drilling fluid under ultra-high temperature and ultra-high pressure

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Abstract

Micro-nano-based drilling fluid has attracted a strong interest due to its attractive properties, and micro-nano composite materials have great potential for developing intelligent drilling fluid. In this study a novel hydrophobic associated polymer based nano-silica composite with core–shell structure was prepared and characterized by PSD, SEM, TEM and ESEM. The results showed that the composite, as a micro-nano drilling fluid additive, possessed excellent properties such as thermal stability, rheology, fluid loss and lubricity. Especially, it could plug the formation effectively and improve the pressure bearing capability of formation significantly.
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Keywords: Composite materials; Polymers; Microstructure; Thermal properties; Petroleum engineering

1. Introduction

As we all know that the drilling fluid in the drilling process of oil and gas well drilling engineering can be seen as the equivalent to the blood in the human body. Commonly used water-based drilling fluid mainly consists of water, bentonite, viscosity increaser, fluid loss reducer, lubricants, etc. But field application shows that it is often impossible to fulfill certain functional tasks that are essential in challenging drilling and production environments using conventional macro-type fluid additives due to their inadequate physical, mechanical, chemical, thermal and environmental characteristics [1]. Therefore, the petroleum industry is looking for physically small, multi-functional, biologically degradable, thermally stable, environmental friendly, novel polymers or natural products for designing intelligent drilling fluids to use virtually in all areas of petroleum exploration and exploitation [2]. Fortunately, with the rapid development of nanotechnology, micro-nanomaterials make drilling fluid with multiple functions simultaneously become possible and using micro-nanomaterials to prepare the high performance intelligent drilling fluid has a greater potential [3–6]. The intelligent drilling fluid can meet the borehole stability, environment-friendly, reservoir protection at the same time, and can identify the different complex circumstances down hole automatically while drilling as well as self-change its function to ensure the drilling engineering smoothly. In this paper, a novel hydrophobic associated polymer based nano-silica composite with core–shell structure, mainly used for developing intelligent drilling fluid additive for drilling ultra-deep well under ultra-high temperature, ultra-high pressure and salinity, was prepared with Acrylamide (Am), 2-Acrylamide-2-methylpropane sulfonic acid (AMPS), Maleic anhydride (MA), Styrene (St) and nano-silica via inverse micro emulsion polymerization and sol–gel preparation.

2. Experimental

2.1. Synthesis

The polymerization of the novel polymer based nano-silica composite with core–shell structure (coded as SDFL) was

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divided into two steps. Firstly, the block hydrophobic associated polymer P(AM–NaAMPS–MA–b-St) was prepared by Acrylamide, 2-Acrylamide-2-methylpropane sulfonic acid, Maleic anhydride and Styrene via inverse micro emulsion polymerization. In brief, 0.06 mol AMPS and 0.01 mol MA were dissolved into 120 mL deionized water, and adjusted pH value to 7 by adding 2 mol/L NaOH at 50 °C, 0.1 mol AM, 0.02 mol St, 3 g span-80 and 1 g OP-10 were added with deionized water to control the total concentration at 15%. Then the mixture was loaded into a reaction flask, stirred in high speed and protected by nitrogen for 30 min. 0.07 g $K_2S_2O_8$ was added and heated up to 90 °C and deoxygenated with nitrogen for 4–5 h, then cooled the reactor to 60 °C. Secondly, the polymer based nano-silica composite was prepared via sol–gel preparation by adding a certain amount of tetraethyl orthosilicate and nano-silica (12 nm, AEROSIL 200, Germany) into the reaction flask to react 2–3 h. Finally, the composite was obtained by filtering, washing and drying.

2.2. Characterization

The particle size distribution and specific surface area were analyzed with ultra-high speed intelligent laser diffraction particle size analyzer delivers (PSD, Malvern Mastersizer 3000, Britain). Scanning electron microscopy (SEM, Hitachi S4800, Japan), transmission electron microscopy (TEM, Hitachi JEM-2100UHR, Japan) and environmental scanning electron microscope (ESEM, Quanta 450, FEI) were used to study the morphology, structure and in situ surface topography of wet samples of the composite. The viscosity of the drilling fluid was tested with six-speed rotational viscometer (ZNN-D6, Qingdao). The rheological and filtration test procedure were executed by the American Petroleum Institute drilling fluid indoor test criteria [7]. The fresh water-based drilling fluid and saturated NaCl-based drilling fluid were prepared according to the report of Yan et al. [8]. Micro-model drilling fluid flooding and drilling fluid displacement experiment of core column were carried out according to the report of Li et al. [9–11]. The resistance factor (R_{es}) and the residual resistance factor (R_{res}) were calculated using the following equations

$$R_{es} = P_p/P_w \tag{1}$$

$$R_{res} = P_s/P_w \tag{2}$$

where P_p, P_s and P_w are the plugging flooding pressure, subsequent water flooding pressure and fresh water-based mud flooding pressure, respectively (MPa).

3. Results and discussion

The PSD curve of SDFL in aqueous solution is shown in Fig. 1(A). One can see that the median particle size of this composite material was 285 nm and the range of PSD was narrow and therefore, it had a very large surface area, reaching to 22.37 m^2/g. The solid fresh sectional SEM image of SDFL in Fig. 1(B) shows that composite morphology was particulate and the particles between each other were connected through the polymer matrix, and PSD was in the range of 280–320 nm. Fig. 1(C) shows the TEM image of SDFL in an aqueous solution, from which we can see that the composite particle size was about 300 nm, and the particles well dispersed in aqueous solution indicating that the hydrophobic associating polymers had been successfully coated on the surface of inorganic nano-silica and a micro-nano composite with "core–shell" structure had formed. The ESEM image of the 0.3% SDFL in aqueous solution was shown in Fig. 1(D). We can see that, the solution emerged with strong cross linking properties after nano-silica introduced into the polymer, and the grid structure was close.

Table 1 lists the influence of SDFL on apparent viscosity (AV), plastic viscosity (PV), yield point (YP), filtration loss (FL_{API}), HTHP filtration loss (FL_{HTHP}) and Lubrication coefficient of the fresh water-based drilling fluid and saturated NaCl-based drilling fluid after aging under 230 °C. As indicated in Table 1, after adding SDFL, drilling fluids displayed a large increase in AV, PV, YP, and a significant decrease in FL and lubrication coefficient. The drilling fluid systems set temperature stability, salt tolerance, perfect rheological properties, good lubricity, and fluid loss control properties in one, and the function of drilling fluids showed significant pluralism. The excellent properties of micro-nano-based drilling fluid were due to the unique molecular design of SDFL in which polymer matrix with high degree branching structure had big steric hindrance and micro-cross-linked structure which combined rigidity and thermal stability of nano-silica with characteristics of hydrophobic associating polymers together. The polymer matrix could provide viscosity at high temperature and the micro-nanoparticles could cause that the entropy of SDFL has a much higher contribution to the specific heat than that of conventional drilling fluid agents, and thereby, when the micro-nanoparticles were circulated into the bottom, more heat could be absorbed to improve the performance of the other systems of drilling fluids. Besides, Si–OH at the surface of the composite could react with Si–OH at the surface of clay by poly-condensation [12] and produce strong chemical adsorption which resulted in thickening hydration membrane and increasing hydration membrane repulsive force at high temperature and high pressure. Thus, the flocculation among clay particles at high temperature and high pressure was prevented effectively to keep a reasonable particle distribution in drilling fluid and form a thin-dense mud cake. Meanwhile, the ultrafine spherical micro-nano particles may form an ultrathin bed similar to the spherical bearing type surface between the pipe and the wellbore [13] and thereby can make easy sliding of the drill string along the spherical bearing surface.

In order to study the plugging ability of micro-pores and micro-cracks and wellbore stability by SDFL, the micro-model drilling fluid flooding and drilling fluid displacement experiment of core column were carried after fracture-making. From Fig. 2, the displacement pressure at two ends of core column by fresh water-based mud flooding was 0.1 MPa, while the displacement pressure by micro-nano composite based mud

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