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Preparation of a micron-size silica-reinforced polymer microsphere and evaluation of its properties as a plugging agent

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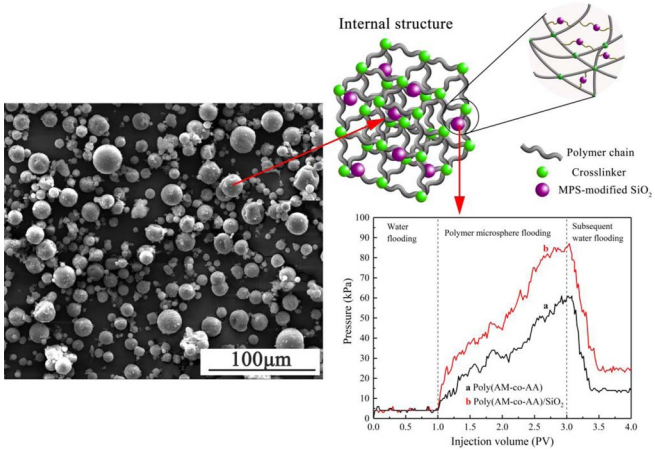
GRAPHICAL ABSTRACT

Polymer microspheres with a reinforcing filler of MPS-modified SiO_2 show better plugging performance.



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ABSTRACT

A kind of silica-reinforced polymer microspheres was proposed and prepared by inverse suspension polymerization, in which the nano-silica modified by 3-(methacrylyloxy) propyl trimethoxysilane (MPS) was used as reinforcing material. Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and dynamic light scattering (DLS) were utilized to analyze their structural properties. The effects of SiO_2 on swelling capability, mechanical shear resistance, stability and viscoelastic property were studied. Plugging behavior was examined by sand pack model experiments. The results show that SiO_2 can effectively adjust swelling behavior, and greatly improve mechanical shear resistance of polymer microspheres. Compared with conventional polyacrylamide microspheres, these silica-reinforced polymer microspheres displayed better dispersion stability and viscoelastic property. In addition, these microspheres exhibited an improved thermal stability, where the maximum degradation temperature was 11 °C higher than that of conventional ones. The plugging rate can be 83.87% and was promoted by 11.54% in sand pack model. This work can provide a viable candidate material for the deep conformance control in oilfields, and supply theoretical support for its application.

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1. Introduction

With the ongoing increase of global economy, the demand for crude oil has grown constantly. However, long-term traditional water flooding in the later stage of oilfield development has aggravated the heterogeneity of reservoirs and fluid mobility differences. As a result, it's easy to form water monolayer breakthrough, resulting in water cone, water channeling, fingering and causing injection water flows inefficient at last [1–3]. This situation triggers that the low-permeability zones can't be swept, and more than 70% of remaining oil is still retained in formation [4]. Thus, improving the sweep efficiency of these areas by plugging the low effective circulation channels is important to enhance ultimate oil recovery in heterogeneous reservoirs [4,5]. To meet that goal, a large amount of plugging materials have been extensively applied in mature oilfields, such as the linked polymer solution, water-based polymer gel and movable weak gel [1,6–9]. However, the cost of the linked polymer solution is high, and its performance is susceptible to harsh reservoir conditions for seriously heterogeneous reservoirs. Furthermore, gel systems have some inherent defects, including uncontrolled gelation time and gelling strength, weak blocking effect, and poor adaptability for high salinity reservoirs [3].

To overcome the problems mentioned above, particle systems were synthesized in surface facilities [10–13]. Among these, polymer microspheres for conformance control have attracted great attentions during past two decades [10,13–18]. Polymer microspheres have excellent properties, such as diverse synthetic methods, controllable particle size, good dispersion stability, swelling capability, and a certain degree of elasticity. Moreover, polymer microspheres are not sensitive to reservoir minerals and formation water salinity [15,16]. The polymer microsphere conformance control technology relies mainly on polymer microspheres to absorb water, plug the pore throat in formation so that making the injected water change the direction [2,16]. Because of microspheres' elastic deformation capacity, they can pass through the pore throat under a large pressure difference to achieve deep dynamic conformance control and oil displacement. However, conventional polyacrylamide microspheres would be easily broken due to the poor shear resistance and complicated formation conditions, and their blocking effect is not significant enough [13,18]. Hence, it is crucial for petroleum industry to develop more excellent polymer nanocomposite microspheres to conquer those problems.

Organic/inorganic composite materials have been studied deeply for a long time [19–25]. These materials combine the advantages of the organic polymer (e.g., flexibility, ductility, and processibility) and the inorganic materials (e.g., rigidity, thermal stability). Moreover, they usually have some unique properties due to the addition of nanofillers. Inorganic nano-materials include nano-metal (e.g., Au, Ag), nano-oxide (e.g., TiO_2, Al_2O_3), semiconductors (e.g., InSb, PbS) and so forth, among which nano-silica is viewed as being very important. Nano-silica has excellent characteristics, such as mechanical, thermal, rheological, and chemical properties. Therefore, it has broad development space in the application of polymeric/inorganic nanocomposite materials. However, nano-silica is easy to aggregate causing it difficult to disperse evenly in the mixed system due to the high surface activity and specific surface area [26,27]. And the performance improvement is limited by physical blending method merely. Thus, surface modification of SiO_2 is urgently needed which can not only effeciently enhance the dispersion performance of SiO_2, but also combine SiO_2 with other materials by chemical bonds (generally C=C) [28,29]. In this way, the performance of composite materials can be improved in practical application [19,27,30–32].

Therefore, this research focuses on the preparation of the silica-reinforced polymer microspheres Poly(AM-co-AA)/SiO_2 and how modified SiO_2 nanoparticles enhance the desired characteristics of polymer microspheres. The structural properties of Poly(AM-co-AA)/SiO_2 were characterized by FTIR, SEM, and DLS. The effects of SiO_2 particles on the properties of Poly(AM-co-AA)/SiO_2, such as swelling properties, mechanical properties, stabilities, and viscoelastic properties were examined in detail. The plugging behavior of Poly(AM-co-AA)/SiO_2 indicated that these composite microspheres can promote the plugging ability and improve the sweep efficiency in complex reservoirs during water flooding, which can enhance flooding effect significantly.

2. Experimental

2.1. Materials

Acrylamide (AM), ammonium persulfate (APS), sodium chloride (NaCl), ammonium chloride (NH_4Cl), anhydrous ethanol (C_2H_5OH), hydrochloric acid (HCl) and polyethylene glycol (PEG-200) were all purchased from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). Silicon dioxide (SiO_2, 30 nm), acrylic acid (AA), and N,N'-methylene bisacrylamide (MBA) were supplied by Aladdin Industrial Corporation (Shanghai, China). 3-(methacrylyloxy) propyl trimethoxysilane (MPS) was obtained from Heowns Biochemical Technology Co., Ltd (Tianjin, China). Sodium hydroxide (NaOH) and ammonium hydroxide ($NH_3{\cdot}H_2O$) were obtained from Xilong Science Co., Ltd (Shantou, China). Sorbitan monostearate (SP-60) was purchased from Shanpu Chemical., Ltd (Shanghai, China). Hydrolyzed polyacrylamide (HPAM, relative molecular mass 17.4×10^6) was supplied by Greatwin Chemical Co., Ltd (Zhibo, China). Aviation kerosene was purchased from market. All reagents were used without any further purification. Deionized water was used for the preparation of all aqueous solution mentioned in this paper.

2.2. Modification of silica

The reaction was conducted in a 250 mL three-necked, round-bottom flask equipped with a mechanical stirrer, a reflux condenser, and nitrogen inlet and outlet. 1 g silica was accurately weighted and then added to the mixture of deionized water and anhydrous ethanol (volume ratio = 1:10) by ultrasonic dispersion. Then the mixture was positioned in the three-neck flask. A certain amount of silane coupling agent MPS was then added dropwise to the system and a few drops of ammonia were added. The flask connected to nitrogen and reflux condenser was placed in a thermostatic water bath at 50 °C. The reaction lasted for 24 h with a stirring speed of 500 r/min. The solution was centrifuged after the reaction was completed. The resulting product was filtered and rinsed several times with anhydrous ethanol and then dried at 60 °C for 48 h. In this way, MPS-modified SiO_2 was obtained. The chemical equation is shown in Fig. 1.

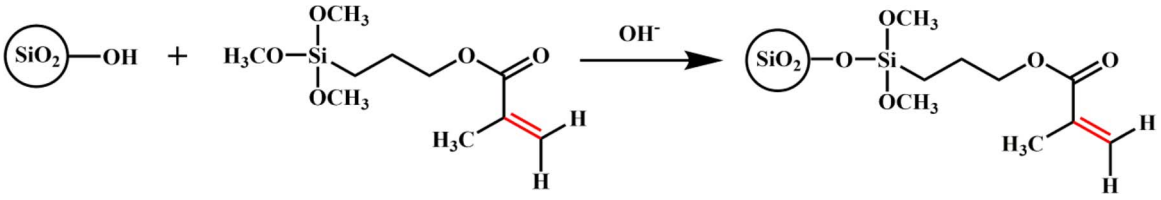

Fig. 1. Synthesis of MPS-modified SiO_2.