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Nanocomposite hydrogels adsorption: Experimental investigation and performance on sandstone core

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ABSTRACT

In this study, adsorption and performance investigation of a $Cr(OAc)_3$/polyacrylamide hydrogel was investigated experimentally to increase hydrocarbon recovery by reducing excessive water. Disproportionate permeability reduction of hydrogel was indicated by inserting in sandstone powder and core. Also, formation of exfoliated microstructure was shown through X-ray diffraction patterns. In order to demonstrate the interaction of nanocomposite (NC) hydrogel and the rock surface as well as its qualitative transformation, infrared spectroscopy was used. Besides, scanning electron microscopy was used to determine the surface characteristics. Based on the results, adsorption decreased continuously and solid/liquid (S/L) ratio increased relatively, while pressure remained independent of any changes. Moreover, the optimum amount of NC hydrogel adsorption was evaluated at the pressure of 2500 psi and S/L ratio of 0.005. The influence of nanoclay concentration on hydrogel adsorption was also studied, using the sandstone core. The adsorption amount was considered about 340 mg/m^2 with maximum adsorption of sandstone core and the NC hydrogel of sulfonated polyacrylamide with 1000 ppm of nanoclay. The capability of hydrogel to increase the reduction of the water relative permeability than oil was more than 12 times. These results confirmed the hydrogel as a proper candidate for field in water shutoff operation.

1. Introduction

Producing high water is an important issue in mature hydrocarbon reservoirs (Vossoughi, 1999). Injecting polymer gel directly in the surroundings of the wellbore is an efficient effort facing this problem. In producing wells, through the reduction of the rock's water relative permeability with respect to oil relative permeability, injection of hydrogel can reduce the water cut. Considering the physical aspect of the issue, the adsorption phenomenon of the water-soluble polymer into the walls could lead to the changing of the permeable medium's two-phase flow properties (Zitha et al., 1998). Adding gels in a petroleum reservoir would lead to a more dramatic drop in permeability of water compared to permeability of oil. This is considered as favorable disproportionate permeability reduction (DPR), and while application of different polymer gel systems leading to DPR is the subject of different studies (Bai et al., 2015; Elsharafi and Bai, 2016; Seright, 2009; Willhite et al., 2000), none of the proposed mechanisms managed to be clearly considered as the primary source of favorable DPR (Zitha et al., 1999). Adsorption is one of the proposed mechanisms of DPR. There are many polymers and gels which could cause a higher decrease in water permeability compared to oil or gas permeability, but extensive field application of the DPR is affected and restricted by different factors. As often synthetic polymer used for polymer gel flooding, partially hydrolyzed polyacrylamide (HPAM) can be degraded at high levels of temperature and salinity, both chemically and thermally (Rashidi et al., 2009). In order to overcome the restrictions, chromium (III)-acetate--sulfonated polyacrylamide pore-filling gels were used (Seright, 2009). Polymer, by adsorbing in porous media, makes a thin layer on the walls of pore and so the effectively obstructs water. However, at the same time oil flow alters slightly (Zitha et al., 1998). In order to apply gel treatment effectively, a substantial amount of adsorption of polymer gel is necessary on the rock surface (Chiappa et al., 1999). Adsorption could be considered as the polymer molecules and the solid surface's mutual interactions, causing the molecules of polymer to stick to the solid surface. It is done mostly by means of physical adsorption which means the creation of the Van der Waal's and hydrogen bonding instead of complete

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chemical bonds formation or chemisorption (Greesh et al., 2008). Basically, the polymer molecules fill the surface's adsorption sites while the surface area directly affects the adsorption amount (Chiappa et al., 1999). Besides, it is only the adsorption that can detach polymer molecules from the bulk solution. This can be achieved by adding a free solid powder, e.g. silica, into the solution and agitating until observing an equilibrium (Cankara, 2005). Having a higher segment density compared with the molecular coil in dilute solution and as a monolayer of molecular coil, polymer molecules would be adsorbed on the porous medium surface, considering the model which defines adsorption as a polymer function (Hirasaki and Pope, 1974). According to another investigation in which static adsorption tests were used, adsorption can be controlled by an electrostatic reaction which could be defined as mutual interactions between the solid surface and polymer molecules (Chiappa et al., 1999). Furthermore, different sulfonated polyacrylamide hydrogels have different adsorption properties which indicates that solid to liquid (S/L) ratio has a great importance in changing the polymer adsorption amount on the kaolinite and silica powder surface (Rashidi et al., 2009). Several different factors could affect the conformation of macromolecules. The first one is pressure through which the adsorption of polymer can be influenced on the solid surface besides the main conformation of polymer chains in the bulk solution. Changing the pressure level would result in the linear dimensions of macromolecules modification related to their development amount. Researches carried out about the adsorption of polyacrylamide on the solid particle surface usually ignore the influence of pressure (Chiappa et al., 1999; Rashidi et al., 2009; Zitha et al., 1998). Wilson (1956) reported that the core effective permeability to oil and water would be lowered, considering a 5000 psi higher pressure level at reservoir temperature in the lab. Also, such a pore size distribution change besides the alteration of the core's relative permeability could be the result of an extra level of pressure, which is able to reduce the porosity of a core by 5% (Ali et al., 1987). Due to their great mechanical, thermal, and other properties that increase their technical worth, mixtures producing from polymer besides a small amount of nanoclay have gained a lot of attention nowadays (Okay and Oppermann, 2007; Saghafi et al., 2016; Tongwa et al., 2013). Having two silica–oxygen tetrahedral sheets which cover an aluminum or magnesium octahedral sheet, montmorillonite (MMT) is a clay that is used very often in polymer–clay nanocomposites preparation. Moreover, some researchers argue that AMPS is a clay modifier (Xu et al., 2003), mentioning its ability of widening the d-spacing between platelets to support their claim, that extend from 1.17 nm (pristine clay) up to 2.1 nm, which of course is related to the AMPS/clay ratio used.

Through crosslinking the sulfonated polyacrylamide/montmorillonite (Na-MMT) clay aqueous solutions with chromium triacetate ($Cr(OAc)_3$), using chromium triacetate as crosslinker, this work is aimed to prepare the nanocomposite type of hydrogels (NC hydrogels). Another investigation was also made, performing static adsorption experiments, to clarify how the reservoir pressure and the S/L ratio can affect the adsorption of NC hydrogels on the powdered sandstone of reservoir rock (as the solid phase). In order to determine the structure of the prepared hydrogel and Fourier transform infrared spectroscopy (FTIR) and to qualitatively demonstrate the existence of interaction between NC hydrogel and the rock surface, the X-ray diffraction (XRD) patterns were analyzed. Also, determination of surface characteristics of the NC hydrogel adsorbed layer on the powdered rock surface was performed by scanning electron microscopy (SEM). The effect of nanoclay content on polymer gel adsorption was studied using five different concentrations of nanoclay. The effect of NC hydrogel adsorption on the hydrogel performance as a DPR agent was also studied by coreflooding experiments on the sandstone core, corresponding to the maximum and minimum hydrogel adsorption in the static adsorption on the sandstone powder.

2. Experimental investigation

2.1. Materials

The average molecular weight and sulfonation degree of the used copolymer of 2-acrylamido-2-methyl-propanesulfonic-acid sodium salt and acrylamide in this study, had been 2 million Dalton and 25%, respectively. It was provided under the trade name of AN125VLM by SNF Co. (France) (Fig. 1-a). At high temperatures, some amide groups of sulfonated polyacrylamide copolymer convert to carboxylate groups. This phenomenon is known as thermal hydrolysis, which cause the structure of AN125VLM at 80 °C containing some carboxylate groups (Fig. 1-b).

Chromium triacetate, purchased from Carlo Erba Co. (Italy), was considered to be used as a crosslinker. Sodium lactate, supplied by Merck Co. (Germany) in the form of a colorless liquid, was also required to act as a retarder. The powdered rock samples of sandstone reservoir used as solid phase with specific surface area of about 0.628 m^2/g determined by using nitrogen adsorption method of Brunauer–Emmett–Teller (BET). Na-montmorillonite was used as nano material with d_{001} interplanar spacing of 12.58°A in XRD analysis, supplied by Advanced Technology (Hangzhou, China). As shown in Fig. 2, the SEM micrograph of Na-MMT particles illustrated the nano size of the used Na-MMT.

Sandstone is a classic, sedimentary rock composed primarily of sand-sized particles. Chemically, the composition of sandstone is silicon dioxide (SiO_2 or silica) oriented in the crystalline form. A silica molecule has a tetrahedral structure having a silicon atom at the center besides four oxygen atoms around it. The silicon atoms of adjacent molecules have links with the surrounding oxygen atoms, creating a continuous network type structure. However, only bulk molecules contain this continuous. Discontinuity can occur at the surface, due to the lack of bonding between two oxygen atoms from each silica molecule, which cause negative charges on the surface (Srisuriyachai, 2008). The

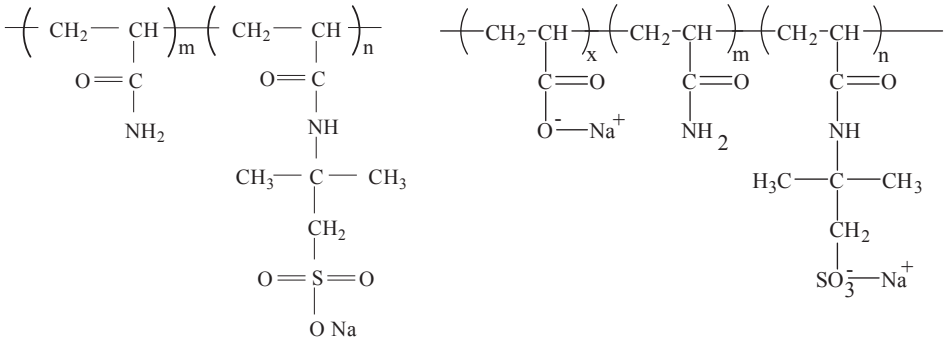

Fig. 1. Copolymer structure of sulfonated polyacrylamide (a: AN125VLM, b: AN125VLM at 80 °C).