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Nanosilica-induced high mechanical strength of nanocomposite hydrogel for killing fluids



Feifei Sun, Meiqin Lin*, Zhaoxia Dong, Juan Zhang, Cheng Wang, Shuanglong Wang, Feifei Song

Institute of Enhanced Oil Recovery, China University of Petroleum, Beijing 102249, PR China

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ABSTRACT

Nano-silica was introduced to enhance the mechanical strength of polymer hydrogels obtained via the crosslinking of polyacrylamide (PAM) and chromium acetate. Rheological properties, compression strength and compressive stress-strain of both nanocomposite and normal hydrogels without nano-silica were investigated by HAKKE rheometer, compression strength test device and electronic universal material testing machine. Moreover, environmental scanning electronic microscopic (ESEM) was adopted to observe the three-dimension network structure of nanocomposite and normal hydrogel, as well as the distribution of nano-silica. The results demonstrated that elastic moduli (G') and viscous moduli (*G*") of nanocomposite hydrogel were both improved with increasing nano-silica concentration. Especially when silica content reached 10 wt%, G' and G" of nanocomposite hydrogel increased over one hundred times higher than those of normal hydrogel. The original compression strength of hydrogel was 70.8 kPa m⁻¹, while the resulting strength of nanocomposite was enhanced to be 196.64 kPa m⁻¹. When the hydrogel were sheared, the normal hydrogel was fractured under low strain, whereas nanocomposite hydrogel was not broken under high strain, and it quickly recovered its original shape after the release of load. In addition, the ESEM images indicated that a large quantity of silica particles aggregated and attached around the polymer chains, and others aggregated to fill into the three-dimension network of hydrogel, which induced the compaction of the space between the network layers and reduced the flowing of free water wrapped in the network, therefore the mechanical strength of hydrogel was enhanced.

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* Corresponding author at: 18# Fuxue Road, Changping, Beijing, PR China. *E-mail address*: 13910509321@163.com (M. Lin).

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1. Introduction

Mature oilfields have been developed into a stage of low production of oil and gas, as well as low pressure due to primary and secondary oil recovery, leading to the difficulties in future exploration. Fortunately, underbalanced drilling and completion technology is not only an excellent method for low pressure reservoir exploration [1–4], but also an advanced reservoir protection technology to reduce lost circulation and eliminate formation damage [5]. During the process of underbalanced operation, the formation pressure is higher than vertical wellbore pressure, which will cause an potential uncontrolled risk of well blowout. Therefore well killing technology that can avoid well blowout has become a key technology for carrying out underbalanced drilling and completion (known as whole-process underbalanced drilling) of reservoir development [3]. Among techniques used for well killing applications, killing fluids is one of the most common and practical methods. Killing fluids containing solid particles and chemicals were ever developed for workover construction and killing operation application, however, the solid particles were potential source of damage to well productivity as well as lost circulation, and the chemicals can react with formation fluids producing harmful precipitation [6–9]. Then a variety of solid-free killing fluids were studied to use in corresponding geological reservoir [9-11], and low-damaging solid-free killing fluids came into research in domestic and overseas [12,13]. But the viscosity of solid-free killing fluids were too low to balance the formation pressure alone, and its lost circulation also caused plugging to low permeability reservoir, so clean isolation fluids that segregated killing fluids from reservoir were developed to increase wellbore pressure, as well as to reduce lost circulation against reservoir damage. Khabibullin et al. [14] used a sort of viscoelastic surfactant fluid as clean isolation fluids to avoid reservoir damage during workover process. Fan et al. [15] mixed solid-free fluids with polyacrylamide solution to increase viscosity and specific gravity of killing fluids, however, the strength of these viscoelastic fluids were still not high enough to resist the formation pressure, and they were broke through easily by formation pressure. Nwoke et al. [16] prepared high-viscosity polymer hydrogel used as killing fluids, which was prepared by water-soluble polymer crosslinking with crosslinkers. Arangath et al. [17] combined solid-free viscoelastic fluids with whole-process underbalanced drilling technology applying in low permeability reservoirs for well drilling and completion to achieve reservoir protection. Recently polymer hydrogel has been still used as killing fluids in certain oilfields, but its application for relative high-pressure reservoir was limited by its low mechanical strength and toughness [18,19]. Because the reaction between crosslinkers and polymer are so random that the crosslinking points are distributed disorderly in the network structure, leading to the polymer chains between crosslinking points in different lengths, once short chains suffered fracture, followed by the crack of entire hydrogel structure. As a result, polymer gel with weak strength was broken through easily by high formation pressure, resulting in the risk of well blowout [20,21]. Therefore, it is significant to prepare high-strength polymer hydrogel for well killing. Inorganic solid nano-silica is featured by its small size, large specific surface, excellent stability, enhancement and thickening properties [22–24], which make it possible to improve the strength and toughness of hydrogel greatly without side reaction. Such types of mechanically strong composite hydrogels have been developed and showed great potential applications in biological fields and industry. For example, silica/polyacrylamide core-shell nanocomposite hydrogels and poly(dimethylacrylamide)/silica hybrid hydrogels were separately synthesized in biomedical and pharmaceutical applications [25,26]. Polyacrylamide (PAM) composite hydrogel containing silica sol using free-radical polymerization was prepared for potential industrial application [27]. PAM/silica composite capsules by inverse Pickering emulsion polymerization was applied for wastewater treatment [28]. Hybrid hydrogel of poly(AM co DADMAC)/silica sol was synthesized for adsorbent in the removal of methyl orange from aqueous solution [29]. Aluminum- and sodium-modified colloidal silica was incorporated into PAM hydrogel for industrial application [30]. However, none of these hydrogels was tried to be applied in oilfield. Aqueous hybrids of silica nanoparticles and hydrolyzed polyacrylamide was ever used for enhanced oil recovery in reservoirs [31], but no chemical or physical crosslinking was reacted between silica and polymer, so the hybrid has high viscosity without mechanical strength. Obviously, it will be of great potential to construct composite hydrogels with high viscoelasticity, compressive strength and adhesion to be applied as killing fluids.

Herein nano-silica was added into polymer/chromium acetate crosslinking systems to form composite hydrogel through coordination polymerization, in which silica can be dispersed into the network of hydrogel because of its small diameter, and develop high-toughness, wear-resisting, and good-stability to enhance mechanical strength and deformation reversibility of nanocomposite hydrogel. In this paper, the mechanical strength including viscoelasticity, compression strength, compressive strain–stress of nanocomposite hydrogel for well killing application were studied, and the distribution of silica particles that induced the enhancement of hydrogel strength was also observed so as to investigate the enhanced mechanism.

2. Experimental

2.1. Materials

Partially hydrolyzed polyacrylamide (HPAM) used with an average molecular weight of 10⁷ and a hydrolysis degree of 13% was purchased from SNF (China) flocculant company in the form of white powder. Thiourea used as stabilizer was purchased from Weifang Chenyang Chemical Co. P.R. China in the form of white powder. Chromium acetate used as crosslinker was purchased from Shanghai branch of Shanxi Xiaxian Yunli Chemical Co. P.R. China in the form of dark green solution. Nano-silica, which is hydrophilic particles of 9–20 nm in size, used as enhancer was purchased from Zhejiang Yuda Chemical P.R. China in the form of white powder.

2.2. Synthesis of nanocomposite hydrogel

The following nanocomposite hydrogels (Table 1) were synthesized according to the following steps. First, nano-silica powder was added to deionized water very slowly under continuous

Tabl	e 1			
Com	position	of	sam	ples.

Sample	HPAM % (mass fraction)	Nano silica % (mass fraction)	Chromium acetate % (mass fraction)
GEL1	2.0	0	0.2
GEL2	2.0	1	0.2
GEL3	2.0	3	0.2
GEL4	2.0	5	0.2
GEL5	2.0	7	0.2
GEL6	2.0	10	0.2
GEL7	3.0	2.0	0.3
GEL8	3.0	4.0	0.3
GEL9	3.0	6.0	0.3
GEL10	3.0	8.0	0.3

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