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Multiple Hydrogen Bonding-Assisted High-Strength Hydrogel of Silica/Polyacrylamide Nanocomposite Cross-Linked with Polyethylenimine

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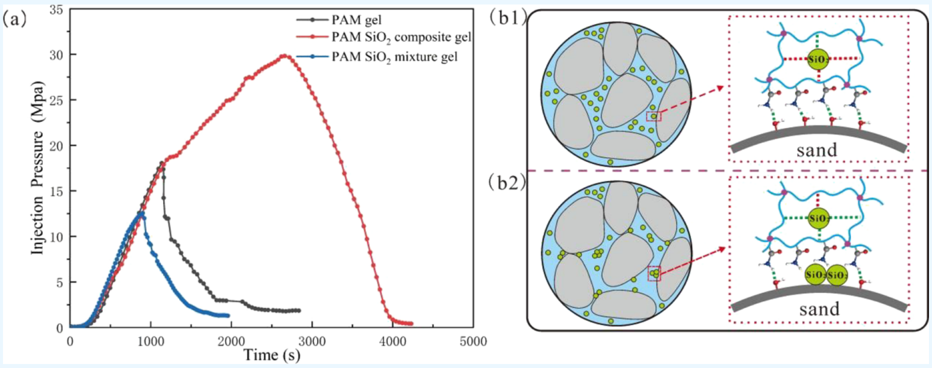

ABSTRACT: The nanocomposite gel system has been successfully applied as a water shutoff agent to enhance oil recovery (EOR) or for plugging to control lost circulation events. In this study, the silica/polyacrylamide nanocomposite was synthesized via in situ free radical polymerization of acrylamide (AM) monomers in the presence of silica nanoparticles. The composite was cross-linked with polyethylenimine to prepare a high-strength hydrogel. The viscosity test was conducted to determine the gelation time of the gel. Rheological measurements and sand pack breakthrough pressure tests were carried out to measure the gel strength. Attenuated total reflectance-Fourier transform infrared (ATR-FTIR) and scanning electron microscopy (SEM) tests were adopted to characterize the structure and morphology of the gel. The results show that compared to polyacrylamide (PAM) gel, the gelation time of the nanocomposite gel will decrease with increasing gel elasticity modulus, and the breakthrough pressure of the nanocomposite gel is 29.82 MPa, which increased by 65%. As shown in the ATR-FTIR test, this can be attributed to the presence of multiple hydrogen bonds for the PAM molecule with both silica and quartz sand particles. In the composite gel, hydrogen bonding mainly forms between the O atoms of PAM and the H atom on the surface of silica, enhancing gel strength and elasticity modulus with more cross-linking density and less porosity. Moreover, H bonding between additional $-NH_2$ of PAM and quartz sand particles helps improve gel plugging pressure. However, in the silica and PAM mixture gel, the H bonding of silica occupies $-NH_2$ of PAM, which became unavailable to attach on the sand surface, reducing the breakthrough pressure by 30%, although it can enhance the rheological strength. This study suggests that in situ composite of silica in PAM can not only greatly improve gel rheological strength but also help maintain the strong adhesion of PAM molecules onto quartz sand, resulting in better plugging performance in the sand reservoir.

1. INTRODUCTION

Hydrogels have great potential applications in the oil industry owing to their biocompatible and environmentally friendly properties.[1−3] One problem gel can address is excess water production in oil fields, which can cause less production of oil and gas and extra cost of billions of dollars to dispose the water.[4−6] The main reason for this problem is reservoir rock heterogeneity. Injected water will tend to enter through the high-permeability zone with oil remaining in the low-permeability rock formations.[7] In situ cross-linked polymer gel was widely used to migrate this problem.[8] The gel system was prepared with high-molecular-weight polymers and cross-

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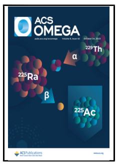

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linkers. Before cross-linking, the gelling solution was pumped in to target zone.[9] After aging for certain periods at reservoir temperature, the three-dimensional structure of gel formed, minimizing water production and increasing the sweep coefficient in the aspect of water shutoff and conformance control.[10−13]

Besides, another problem that gel can address is lost circulation (LC), which is a common issue during drilling operations.[14] Lost circulation is prone to happen in formations including pore throats, induced or natural fractures, and caverns.[15] Events such as a blowout may take place in severe situations. Cross-linked polymers can seal a large scale of pore space or fracture and offer an efficient solution to deal with severe lost circulation events.[16] Hashmat investigated the gel system composed of hydrolyzed polyacrylamide, and polyethylenimine was taken as a cross-linker.[17] The polymeric systems can act as loss prevention and sealing agents. Soliman designed an organic cross-link polymer system.[18] Together with plugging materials, severe loss of circulation events can be cured. High-molecular-weight synthetic polymers used in the gel include polymers or copolymers of acrylamide (AM) monomers, poly(vinyl alcohol), polyacrylonitrile (PAN), etc. Cross-linkers can be divided into inorganic and organic cross-linkers.[4] Among all the cross-linkers, PEI is environmentally friendly and therefore widely used.[5,19]

To improve the hydrogel mechanical strength, many methods have been proposed. Among them, nanocomposite hydrogels (NC gels) exhibit advantages of easy modification and simple preparation. Nanoparticles are added into the gel and can have cross-linking interaction with polymers through physical force or covalent bonding. The commonly used type of nanoparticle is silica. On the surface of silica contains large amounts of −OH groups, which can link to other molecules chemically or physically. Moreover, hydrogen bonding can form between these −OH groups and the polymer chains of hydrogels. Therefore, the silica can be used as a physical cross-linker within NC gels to enhance the gel.[20−23] Silica nanoparticles were directly mixed in PAM gelling solution by Liu.[24] Hydrogen bonds and the electrostatic interaction of the silica can increase the gel strength. Jia developed a highly stretchable nanocomposite hydrogel.[20] According to FTIR test results, the higher strength of the gel can be attributed to hydrogen bonds of PAM polymer chains with silica nanoparticles. Silica functionalized or mixed with hexadecyl trimethylammonium bromide can further enhance gel strength.[25,26] After adding the functionalized silica, the gel breakthrough pressure can increase by 13.4%.[25] However, silica functionalization requires more complex procedures and becomes costly while the effect of directly mixing silica during gel preparation is not remarkable due to lack of full dispersion. Then, how to apply hydrogen bonds of nanosilica to enhance in situ cross-linked hydrogel requires further investigation.

In this study, by free radical polymerization of AM monomers in the presence of silica nanoparticles, a new nanocomposite was synthesized. The composite was then cross-linked with PEI to prepare H-bonding-assisted high-strength hydrogels. The gelation time and mechanical properties of nanocomposite gel were studied. Based on SEM and ATR-FTIR test results, the effect of H bonding types on gel strength was discussed.

2. EXPERIMENTAL SECTION

2.1. Materials. The silica nanoparticles (SiO_2 NPs, 30 nm), the cross-linker polyethylenimine (PEI, M_w = 70,000 g/mol), sulfuric acid, sodium formate, carbamide, and the initiators including ammonium persulfate (APS), potassium metabisulfite (PMS), azodiisobutyronitrile were used as received. Acrylamide (AM) was purified by recrystallization before used. All of the chemicals were of analytical grade and purchased from Aladdin Chemical Reagents, Inc. All aqueous solutions in the experiments of this study were prepared with deionized water.

2.2. Methods. *2.2.1. Synthesis of the PAM-SiO_2 Composite.* The PAM-SiO_2 composite was prepared via radical polymerization. 0.1 g of SiO_2 nanoparticles was dispersed with 20 mL of deionized water and stirred with a magnetic stirrer at about 200 rpm. Then, it was put under ultrasonic dispersion for 0.5 h. AM (0.25 mol), ammonium hydroxide (0.0052 mol), sodium formate (0.0006 mol), and carbamide (0.001 mol) were sequentially dissolved in 80 mL of deionized water and then mixed with SiO_2 dispersion. The pH of the mixture was adjusted to 10.0 with sulfuric acid. To get a homogeneous solution, stirring was continued at 500 rpm for 30 min at 1 °C, with bubbling nitrogen to deoxygenate at the same time. The initiators including 0.0003 mol of APS, 0.0003 mol of PMS, and 0.002 mol azodiisobutyronitrile were added into the mixture and stirred at 20 °C for 4 h to obtain the composite. Then, the sample was kept at room temperature (25 °C) for another 12 h to promote the conversion rates.[27]

PAM samples with various molecular weights were prepared by changing the AM monomer concentration (1.8, 2.5, 3.2, and 3.9 M), following the same procedures as those of the PAM-SiO_2 composite without the addition of nanoparticles, and the temperature was set at 5 °C during the reaction.

In order to remove the residual mobile monomers or ions, the prepared PAM and PAM-SiO_2 composites were put in dialysis bags and immersed in a large excess of water. Water was changed every day until the conductivity of the dialysis solution was constant. Using an Ullman viscometer with the one-point method,[28] intrinsic viscosity of the PAM-SiO_2 composite was determined to be 693.20. PAM synthesized with different AM monomer concentrations were named PAM-L, PAM-MI, PAM-MII, and PAM-H, whose intrinsic viscosities were 626.48, 632.99, 741.48, and 859.77, respectively.

2.2.2. Preparation of the Hydrogel. PAM-SiO_2 mixture gel preparation. First, 0.2 g of SiO_2 NPs was added into 20 mL of neutral deionized water, stirring for 20 min to obtain a dispersion. The dispersion was then ultrasonic dispersed for 0.5 h. The PAM solution with a mass fraction of 2% was prepared by diluting the synthesized PAM sample. Then, SiO_2 dispersion was added into the PAM solution drop by drop, stirring for 0.5 h to get a homogeneous solution. PEI with 0.3% mass fraction was added to the solution and stirred for another 10 min. Thereafter, the gelling solution was put into a polytetrafluoroethylene bottle. The gel cross-linking began when the bottle was put in a 30 °C water bath or oven, and the reaction lasted for 72 h at maximum.

Following the same steps, PAM gels were prepared without the addition of SiO_2 NPs. The PAM-SiO_2 composite gels were obtained following the same procedure as that for the PAM gel, with the PAM sample replaced by the nanocomposite.

2.3. Characterization. *2.3.1. Gelation Time and Gel Rheological Measurements.* The gelation time is the amount of time it takes for the mixture to transform in the liquid form to a gel. A Brookfield rheometer (DVNEXT model, Brookfield, Middleboro, MA, U.S.A.) was used to measure the viscosity change with time. Approximately 180 mL of the gel sample was put in a polytetrafluoroethylene bottle with a capacity of 200 mL. The Brookfield viscometer was preheated, and then, the bottle was put under it, with the spindle of the viscometer inserted into the gel. The gelling solution was stirred at a rate of 1 rpm and its viscosity was recorded every 120 s at 30 °C. The inflection point on the plot of the viscosity time is defined as the gel point.

The rheological properties test of the gel were carried out at 30 °C by a rheometer (Anton Paar MCR302X, Anton Paar Co. LTD, Austria). The sweep frequency was in the range of 1−10 Hz.

2.3.2. Morphological and Structural Characterization of Hydrogels. The surface morphology observation of the hydrogel was performed using a FEI Nova Nano SEM 450 instrument (Thermo Fisher, America) at a voltage of 15 kV. Before the test, gel samples were freeze-dried under vacuum. Then, the sample was cut into pieces and coated with Au−Pd alloy in a sputter coater under an argon atmosphere.

The ATR-FTIR spectra of the hydrogel were measured with a Nicolet IS50 infrared spectrometer (Thermo Fisher, America) over the scanning range between 4000 and 500 cm^{-1} with a resolution of 4 cm^{-1} and averaged 32 scans at room temperature.

2.3.3. Gel Breakthrough Pressure Measurements. The mechanical strength of the hydrogel can be characterized by measuring breakthrough pressure with sandpack flooding tests.[29] Figure 1 shows the sandpack flow apparatus setup. First, the sandpack was filled with 16−26 mesh quartz sand with a permeability of about 3000 mD, and then, the sand was put into the oven at a temperature of 30 °C. The sandpack was saturated with deionized water at a flow rate of 30 mL/min. The pressure differential between two ends of the sandpack was recorded and the permeability was calculated according to Darcy's theorem. Then, 8 pore volumes of gelling solution are injected into the sandpack at a flow rate of 1 mL/min. After the gelation is finished in 72 h, deionized water is injected into the sandpack at 1 mL/min, and the repulsion pressure in the injection end is recorded and the maximum pressure is referred to be gel breakthrough pressure.

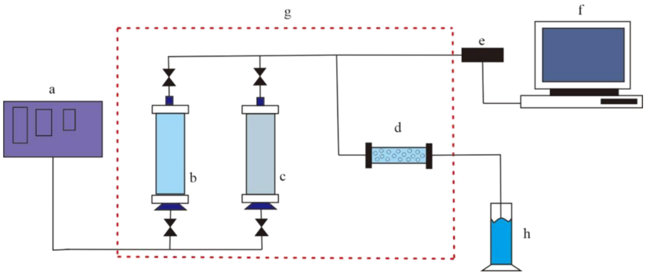

Figure 1. Apparatus setup for breakthrough pressure test ((a) pump, (b) water, (c) polymer gel, (d) sand pack, (e) pressure gauge, (f) computer, (g) hot air oven, (h) volumetric cylinder).

3. RESULTS AND DISCUSSION

3.1. Gelation Time of Hydrogels.

As can be shown in Figure 2a, with the intrinsic viscosity of PAM increasing, gelation time will decrease from 64.8 h of PAM-L gelling solution to 42.3, 31.3, and 25.7 h for gelling solutions of PAM-MI, PAM-MII, and PAM-H, respectively. The cross-linking reaction between PEI and PAM is considered to occur between imine nitrogens on PEI and amide groups on the PAM.[30] With higher intrinsic viscosity, PAM polymer chains are longer, with more carbonyl carbon linked by the amide group. The more cross-linking points on the polymer make them easier to be captured by the cross-linker, resulting in a more nucleophilic substitution reaction rate and less gelation time. The gelation time is 6.0 and 6.5 h for gelling solutions of PAM SiO_2 composite and PAM SiO_2 mixture, respectively. Compared to the PAM gel, gelation times of the composite gel and mixture gel greatly decreased. This can be attributed to silica, which can act as a versatile cross-linker in the system. A large number of nanoparticles are attached to polymer chains, increasing cross-linking points due to the interaction of the −OH groups on SiO_2 with the groups of polymer molecules.[24,25] Although the intrinsic viscosity of the PAM SiO_2 composite is less than that of PAM-MI and PAM-H, gelation time still decreases. As proposed in Figure 2i1, during the preparation of the composite gel, the silica nanoparticles were attached to AM monomers and then remained in the gel, acting as additional cross-linking points to accelerate the cross-linking process. In the mixture gel (Figure 2i2), silica nanoparticles are doped in polymer coils of the gel and accumulate together, which promotes the gelation process and reduces the gelation time.

3.2. Rheological Behavior of Hydrogels.

To further investigate the rheological behavior of hydrogels, the storage modulus (G') and loss modulus (G'') of samples were measured. The storage modulus corresponds to the energy consumed during elastic and reversible deformation. Loss modulus is the amount of energy lost during viscous and irreversible deformation. Storage modulus/loss modulus represents the viscoelastic properties of the gel. In the solidlike material, there will be higher storage modulus than the loss modulus. Instead, for the material with less storage modulus than the loss modulus, it is liquid-like.[31,32] Higher storage modulus can provide strong support and damping effects, thus enhancing the invasive and shear resistance. Therefore, the gel bulk strength can be determined with the storage modulus. As shown in Figure 2b,c, both G' and G'' increased with the frequency, and G' was greater than G'' in the whole scanning frequency range, indicating that all hydrogels displayed a predominantly solidlike behavior. In addition, G' and G'' of PAM gel with higher viscosity (PAM-H, PAM-MII) is lower than that of the rest of the PAM gel samples. Increase of viscosity can reduce water activity and diffusion of PEI molecules, resulting in less cross-linking reaction or density of the gel. Compared with PAM gel, the G' of both composite gel and mixture gel were higher, indicating that silica addition into the gel can improve bulk gel strength. In the two gels with silica, composite hydrogels displayed a higher modulus, revealing that an in situ composite of SiO_2 is a better way to enhance the gel (Figure 2b). Moreover, the G'' of composite gel increases more obviously than the rest as the frequency increases, showing its increasing dynamic viscosity at a higher frequency.

Overall, silica nanoparticles greatly enhanced both elasticity and viscosity of the gel, and more strength enhancements were realized by in situ composite of silica.

3.3. SEM Micrographs of Hydrogels.

The microscopic structures of the gel samples were observed through SEM at room temperature. Figure 2d−f shows the SEM micrographs of the gel samples. As can be observed, when silica was introduced, the porosity greatly reduced with more cross-linking density in the composite or mixture gel. At the same time, silica filled the pores between polymer chains, thereby enhancing the strength of hydrogels. The more gel cross-linking points can induce a higher reaction rate and shorter gelation time, as discussed in Section 3.1. In the images of the mixture gel (Figure 2f2), large-

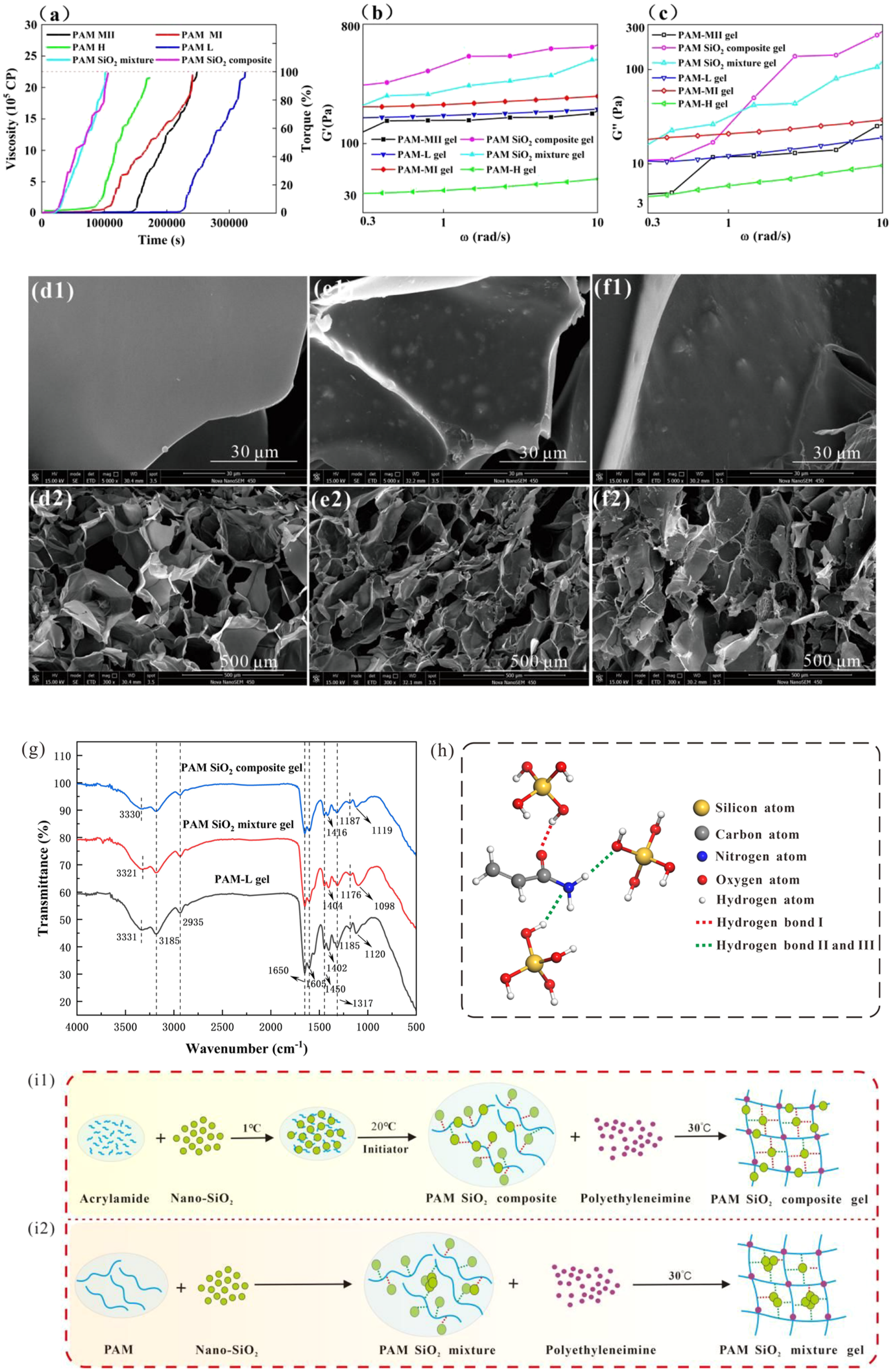

Figure 2. Gel viscosity changes with time (a); storage modulus (G') and loss modulus (G'') of gel samples (b, c); SEM micrographs of PAM-L gel (d1, d2), PAM SiO_2 composite gel (e1, e2), PAM SiO_2 mixture gel (f1, f2); ATR-FTIR spectra of hydrogels (g); types of hydrogen bond in hydrogels (h); proposed interactions of silica nanoparticles in composite gel (i1) and mixture gel (i2).

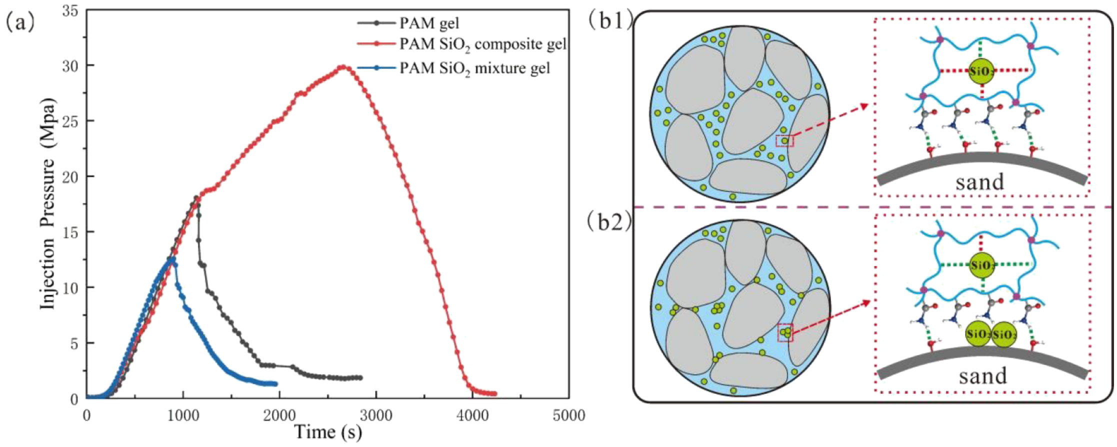

Figure 3. Curve of injection pressure with time (a) and proposed interaction between quartz sand particles and nanocomposite hydrogel (b1) or mixture gel (b2).

scale aggregation of silica nanoparticles can be observed while the nanoparticles in the composite gel are more evenly distributed, which contributes to more intense three-dimensional network structures of the composite gel and more efficient strength enhancing.

3.4. ATR-FTIR Characterization of Hydrogels.

The structures of gel samples were characterized by ATR-FTIR spectroscopy (Figure 1g). In the spectrum of PAM gel, the characteristic bands include $-CH_2-$ (2935, 1448, and 1402 cm^{-1}), C=O (1650 cm^{-1}), −CN− (1317 and 1185 cm^{-1}), and $-NH_2$ (3331, 3185, and 1605 cm^{-1}). In the spectrum of the PAM SiO_2 mixture gel, the corresponding stretching vibration modes of $-CH_2-$ were found at 2933, 1447, and 1404 cm^{-1}. The peak at 1650 cm^{-1} can be assigned to the C=O group with a stretching vibration. Other characteristic bands originating from PAM were also observed, such as −CN− (1314 and 1176 cm^{-1}) and $-NH_2$ (3321, 3184, and 1605 cm^{-1}). PAM SiO_2 composite gel has the following characteristic peaks: 2934 and 1416 cm^{-1} (stretching vibration modes of $-CH_2-$), 1650 cm^{-1} (C=O group with stretching vibration), 1319 and 1187 cm^{-1} (the stretching vibration of C−N), and 3330, 3185, and 1604 cm^{-1} (asymmetric stretching vibration of the NH_2 group).

Compared to that of PAM gel, the stretching vibration of -NH shifted from 3331 to 3321 cm^{-1} in the mixture gel, which can be attributed to hydrogen bonding between the O atom of −OH on silica and the $-NH_2$ group in PAM.[33] In the composite gel, no obvious shift of -NH stretching vibration bands occurs, suggesting that little H bonding forms between the $-NH_2$ group in PAM and silica. As shown in Figure 1h, there are three possible types of hydrogen bonds formed between PAM polymer chains and silica: hydrogen bonds formed between the O atom of the amide groups ($-CONH_2$) and the H atom of −OH on silica (hydrogen bond I), between the N atom of $-CONH_2$ and the H atom of −OH on silica (hydrogen bond II) or between H atom of $-CONH_2$ and O atom of −OH on silica (hydrogen bond III). During the preparation process of the composite gel, all of the hydrogen bonding types should exist in the mixture of AM monomers and silica. After radical polymerization and cross-linking, according to the ATR-FTIR test results, hydrogen bonds I and II rarely remained. As can be inferred, hydrogen bond I dominates in the composite gel. On the other hand, in the mixture gel, there are large amounts of hydrogen bond II or III. This is mainly because H or N atoms of $-CONH_2$ were first captured by silica in the high-viscosity PAM solution during mixing. A few collisions can happen between silica and the O atom within polymer chains, reducing the formation of hydrogen bond I. All hydrogen bonding can enhance gel cross-linking density and strength, and the effect of hydrogen bond I is much better.

3.5. Gel Breakthrough Pressure Test.

In the sand pack flooding tests, PAM-L was used to prepare PAM gel due to longer gelation time and high gel strength. The breakthrough pressures of the composite gel and the PAM-L silica mixture gel were also measured. As shown in Figure 3a, the breakthrough pressure of PAM gel, composite gel, and mixture gel was 18.05, 29.82, and 12.58 MPa, respectively. Compared to PAM gel, the in situ composite of silica in PAM enhanced gel strength by 65%, while for PAM silica mixture gel, breakthrough pressure was reduced by 30%. This can be attributed to the H-bonding interaction between the quartz sand particles and the gel. As proposed in Figure 3b1,b2, in the composite gel, additional H and N atoms of $-CONH_2$ in PAM chains were free to form H bonding (II or III) with sand, enhancing the gel plugging performance. However, silica in the mixture gel occupies the H bonding position, and no more H and N atoms of $-CONH_2$ were available to attach onto the sand, which caused the reduction of gel breakthrough pressure, although it can enhance the strength of bulk gel.

4. CONCLUSIONS

The PAM SiO_2 nanocomposite was synthesized by radical polymerization of AM monomers in the presence of SiO_2 nanoparticles. The composite was used to prepare high-strength gel with PEI as a cross-linker. The following conclusions can be drawn from the study.

(1) SiO_2 nanoparticles in gel can decrease gelation time and enhance its elasticity and viscosity. Compared to directly mixing silica in gelling solutions, an in situ composite of silica in PAM has a better effect to enhance the gel bulk strength.

(2) Compared to PAM gel, the breakthrough pressure of composite gel increased by 65%, indicating that the nanocomposite can greatly improve the plugging performance of gel in sand reservoirs.

(3) The improvements in strength and plugging performance of the composite gel can be attributed to the synergistic effect of multiple hydrogen bonding. The H bonding between O atoms of the amide groups ($-CONH_2$) and H atoms of hydroxyl groups (−OH) on silica can result in more cross-linking density

and higher gel strength. Additional H and N atoms of $-CONH_2$ in PAM chains were free to form H bonds with sand, enhancing the gel plugging performance.

(4) The mixture of silica in PAM solution has a detrimental effect on the gel plugging performance, although it can enhance the bulk strength. This is because H and N atoms of $-CONH_2$ were captured by silica during mixing and became unavailable to attach to the sand, causing less adhesion of gel on the sand particles.

This study proposed a high strength nanocomposite gel system, which was a promising material for water shutoff or lost circulation control. To assess the conditions for nanocomposite gel application in oil and gas fields, the effects of factors, such as temperature, salinity, and SiO_2 concentration, on gel performance will be investigated in future.

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Notes

The authors declare no competing financial interest.

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