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Application of Solvent-Assisted Dual-Network Hydrogel in Water-Based Drilling Fluid for Lost Circulation Treatment in Fractured Formation

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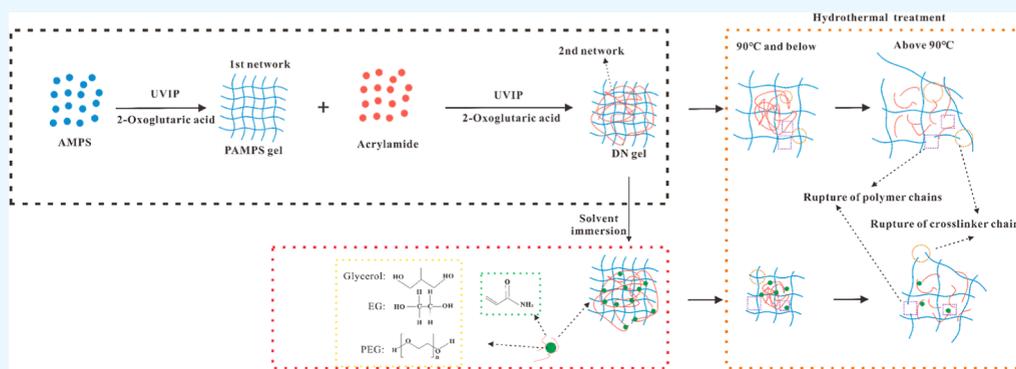
ABSTRACT: During oil and gas well construction, lost circulation caused substantial nonoperation time and extra costs, and hydrogel, resilient and environmentally friendly, was one of the major types of material for lost circulation treatment. To migrate the weak bonding and hydrothermal degradation of conventional single network hydrogels, dual network (DN) hydrogel was prepared and immersed in solvents of polyethylene glycol (PEG), ethylene glycol, and glycerol. The swelling of DN gels at different temperatures was studied with water content and swelling rate tests, and the gel structural and morphology was characterized with attenuated total reflectance infrared spectroscopy (ATR-IR) and scanning electron microscopy test. Then, the compression test and fracture plugging performance test were conducted to study the strength of the gel. The results show that compared to those in ethylene glycol and glycerin, DN gel after immersion in PEG (DN-PEG) exhibits greater compression strength and better plugging performance even at high temperatures. The compression strength of DN-PEG was twice that of DN hydrogel before immersion, and its fracture plug breaking pressure can reach over 10.0 MPa. After undergoing hydrothermal treatment at 90 °C, the compression strength of the DN-PEG was nearly 20 times that of the DN hydrogel, and the fracture plug breaking pressure was still 2.81 MPa. According to ATR-IR spectroscopy, as the molecular weight of the solvent increases, more hydroxyl groups in the PEG have better ability to bind with hydrogen bonds, which greatly inhibits the swelling and polymer chain breakage, thereby reducing hydrothermal degradation in the strength of the dual-network hydrogel. Our work proposed an effective method to reduce the degradation of hydrogel in water at high temperature, and the prepared DN-PEG hydrogel was a promising material for lost circulation treatments in fractured formation.

1. INTRODUCTION

Hydrogel is a kind of polymer network structure material whose main component is water (50−90%). It has a relatively stable physical and chemical structure and presents a three-dimensional network structure in space, which makes the hydrogel resilient and environmentally friendly. During oil and gas production, hydrogels have been widely used in various stages of operation, such as water flowing profile control in oil recovery enhancing and lost circulation treatment during oil and gas well construction.[1,2] Lost circulation occurs mainly due to the presence of naturally fractures, caverns, or highly permeable pore space in formations, along with part or total loss of drilling fluid or cement slurries into the formation. Reportedly, 20−25% of the world's oil fields experienced drilling fluid leakage during drilling operations.[3,4] The loss of drilling fluids can cause severe accidents such as loss of well control, instability of borehole, stuck pipes, and formation

damage, which will lead to more nonoperation time and cost millions of dollars.[5−7] Particle gels are one type of hydrogel widely used to solve this problem. Mohammed et al. developed a low-temperature recrosslinkable preformed particle gel material to control the circulating leakage of drilling fluid.[8] This gel particle has a good plugging effect. When the gel particles were plugged with a 2.00 mm fracture, its sealing pressure can reach up to 1381 psi/ft at room temperature. Zhao et al. reported a self-healing hydrophobic association gel used as lost circulation material.[9] This gel has good mechanical strength, and its plug breaking pressure (PBP) can reach up to 3.5 MPa in a 1 mm fracture. Due to single cross-linking network, the conventional particle gels exhibit low strength. To enhance the mechanical strength of hydrogels, various methods have emerged, such as dual network (DN) hydrogels and nanocomposite hydrogels.[10,11] Nanocomposite hydrogel was proposed to enhance gel strength by adding reinforcing fillers such as clay, cellulose, carbon nanotubes, and nanosilica. Sun et al. added nanosilica into a composite gel system, which consists of polyacrylamide, polyethyleneimine, and cetyltrimethylammonium bromide.[10] The gel strength increased by nearly 10 times than that without nanosilicas. Bai et al. prepared a high strength gel, using acrylamide as monomer and active vinyl group as a cross-linker and ammonium persulfate as an initiator, adding laponite as a physical filler.[11] The polymer gel has a high G' and G'' of more than 1000 and 200 Pa, respectively. Dual network hydrogels can be prepared by physical cross-linking or chemical cross-linking. Sarmah et al. synthesized a physically cross-linked starch/hydrophobically associated poly(acrylamide) self-healing mechanically strong hydrogel.[12] This hydrogel is made of novel starch and acrylamide by physical cross-linking and has the characteristic of maintaining toughness after swelling. The hydrogel also has good compression strength, with a maximum compression strength of 0.034 MPa when the gel compression ratio reaches 90%. The chemical cross-linking DN hydrogels, proposed by Gong et al., have special network structures composed of two polymers with contrasting physical properties.[13] It consists of a neutral polymer as the first network (ductility) and a large number of cross-linked polyelectrolytes (rigid skeleton) as the second network. This dual-network structure has been shown to have significantly improved mechanical properties compared to single-mesh gels, where tearing energies are 10−1000 times higher.[14−16] To further enhance the strength of DN gel, PEG, EG, and glycerol were used by Gong et al. to replace water molecules in the gel to enhance the polymer or cross-linking chains.[17] Lv et al. reported a Gelatin/PAM double network hydrogels with supercompressibility.[18] They compared the compression strength of the double-network hydrogels synthesized by different gelatin concentrations, and when the gelatin concentration was 15%, the compression strength of the double-network hydrogel was the highest, reaching 50.69 MPa. Bai et al. reported dual-network self-healing hydrogels for lost circulation treatment.[19] The first network is composed of sodium alginate and FeCl$_3$·6H$_2$O, while the second network is acrylamide and lauryl methacrylate, using ammonium persulfate as an initiator. The gel tensile stress can reach 0.67 MPa, and its fracture breakthrough pressure is as high as 3.45 MPa at room temperature. Compared to the nanocomposite hydrogels, dual-network hydrogel has better mechanical properties. However, during drilling deep formations, high-temperature conditions is a big challenge for DN gel, where polymer hydrogels suffer from hydrothermal

degradation[20,21] and the method to improve gel strength at high temperature was rarely reported. In this paper, the effect of hydrothermal treatment and solvent exchange on DN gel strength and fracture plugging performance was investigated, and the methods to improve the DN gel high temperature property are explored. Water content and swelling property of DN gels at different temperatures were measured, and attenuated total reflectance infrared spectroscopy (ATR-IR) and scanning electron microscopy (SEM) test were conducted to characterize their structural and morphology. Then, to study the strength of the gel after hydrothermal treatment, the compression test and fracture plugging performance test were carried out.

2. EXPERIMENTAL SECTION

2.1. Materials. 2-Acrylamino-2-methylpropanesulfonic acid (AMPS) is the monomer for the first network of gel, while acrylamide (AM) is the monomer for the second network. N,N′-Methylenebis(acrylamide) (MBAA) is used as a cross-linker, and the radical initiator is 2-oxoglutaric acid. These agents were provided by Aladdin Biochemical Technology Co., Ltd. (Shanghai, China). Ethylene glycol (EG) is purchased from Qiangsheng Functional Chemical Co., Ltd. (Jiangsu, China). Polyethylene glycol (PEG), whose molecular weight is 200 g/mol (PEG-200), and glycerol were purchased from Lingfeng Chemical Reacent CO., Ltd. (Shanghai, China). All of the chemicals were of analytical pure grade and used as received. Deionized water was used to prepare solutions.

2.2. Synthesis of Dual-Network Gels. DN gels are synthesized by radical polymerization in a sequentially two-step process. First, 1 M AMPS solution was prepared and then 4 M cross-linker of MBAA and 0.1 M 2-oxoglutaric acid were added with a magnetic stirrer for stirring. After 30 min of pure nitrogen bubbling, the solution was poured into a 5 mL ampule and put under irradiation of UV lamps (six black lights, 15 W and 365 nm peak). The lights were kept 10 cm away from the ampule, and the photopolymerization lasts for 12 h. Second, the PAMPS was taken out of the ampule and immersed in 2 M Am solution, which contains 0.1 M 2-oxoglutaric acid and 0.25 M MBAA.[22] After at least 1 day, equilibrium is reached, and the sample is taken out and put under irradiation with black lights for another 12 h. The second network cross-linked within the first network and the DN gel was obtained.[23]

The gel sample was immersed in PEG-200, EG, or glycerol, named, DN-PEG, DN-EG, and DN-G, respectively. The gel was weighed every 24 h, and the solvent was replaced every 3 days until swelling equilibrium.

2.3. Hydrothermal Treatment and Swelling Test. The hydrogel samples before or after immersion were cut into 10 × 10 × 10 mm cubes, weighed, and put into a PTFE tank filled with deionized water, loaded into an iron can, and put into an oven at temperature of 25 °C, 50 °C, and 90 or 120 °C. Samples after hydrothermal treatment for 1, 3, 6, 12, or 24 h were taken out and weighed after cooling, and the weight was recorded as m_e. Then, the samples were air-dried at 105 °C until weight remained constant, and the weight was recorded as m_P. Eqs 1 and 2 were used to calculate the hydrogel water content (ω) and swelling rate, respectively.[24,25]

$$\omega = \frac{m_e}{m_P} \times 100\% \tag{1}$$
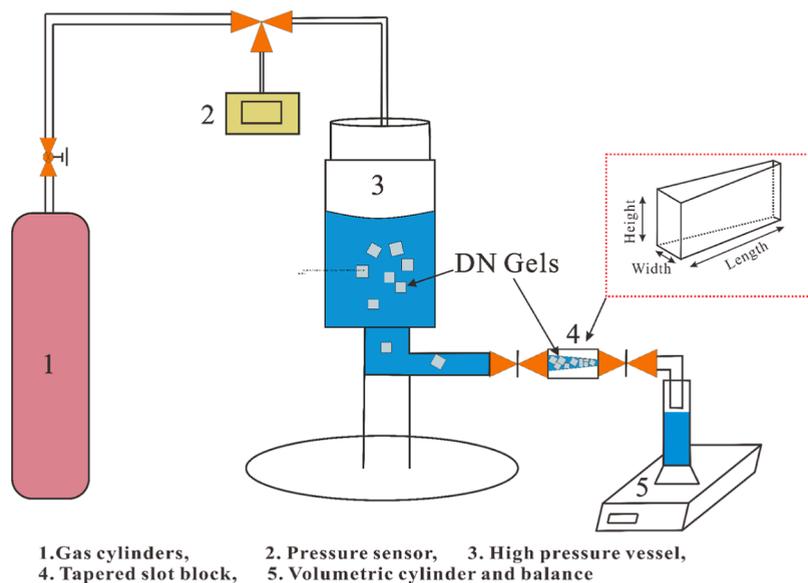
Figure 1. Schematic chart of the PBP test setup.
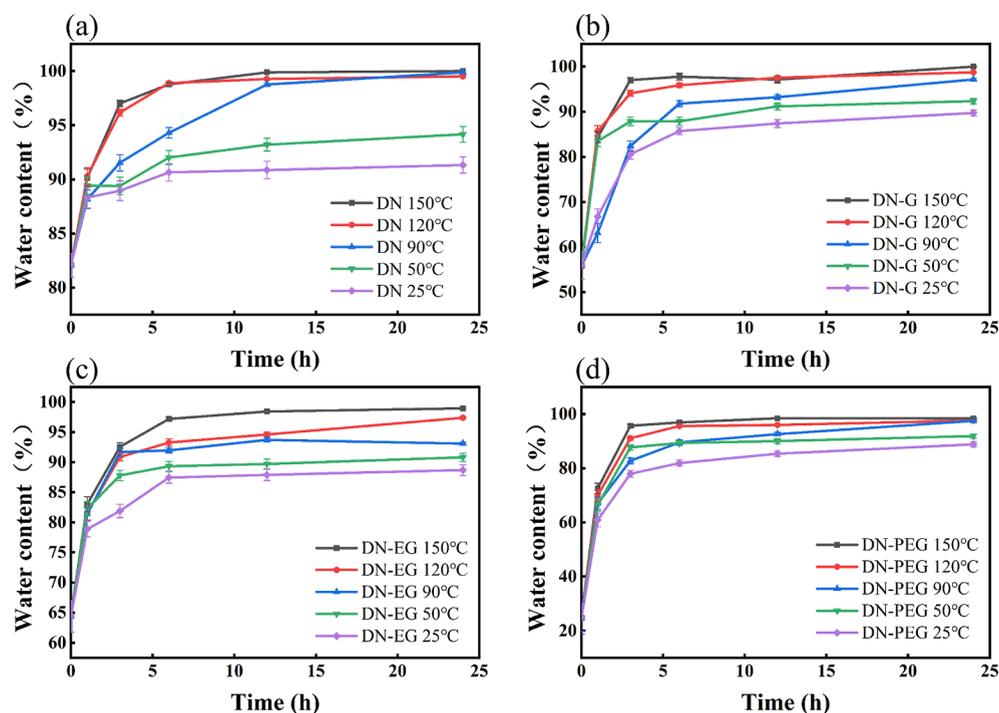
Figure 2. Water content at various temperatures of DN (a) and DN-G (b), DN-EG (c), and DN-PEG (d).

$$\text{Swelling rate}(\%) = \frac{m_e - m_P}{m_P} \times 100\% \qquad (2)$$

2.4. Structural and Morphological Characterization of Hydrogels. To observe the DN gel structure before and after hydrothermal treatments, an SEM test was conducted. The DN samples after hydrothermal treatment were freeze-dried for 72 h. A piece of the freeze-dried samples was cut and its surface coated with Au−Pd alloy under an argon atmosphere. Then, the surface morphology of the sample was examined using an FEI Nova Nano SEM 450 scanning electron microscope (Thermo Fisher, America).

To clarify the change of functional groups in the dual network hydrogel after immersion or after hydrothermal treatment, an ATR-IR spectra measurement was conducted on a Nicolet IS50 (Thermo Fisher, America) at room temperature. Before test, the DN gels were placed in excessive water removing the remaining monomers or ions.[26] The rest of the hydrogels after immersion were used as prepared. The scanning ranged from 4000 to 500 cm^{-1} and the resolution is 4 cm^{-1}. 32 more scans were conducted for each sample in average.

2.5. Compression Strength Test. During the compression test, hydrogel samples are cut into cylindrical shapes (12 ± 0.5 mm high and 28 ± 0.5 mm in diameter), as required by GB/T 7757-93, and the samples were under compression in a servo controlled hydraulic loading device (SHM-60T, China) at loading rate of 2 mm/min.
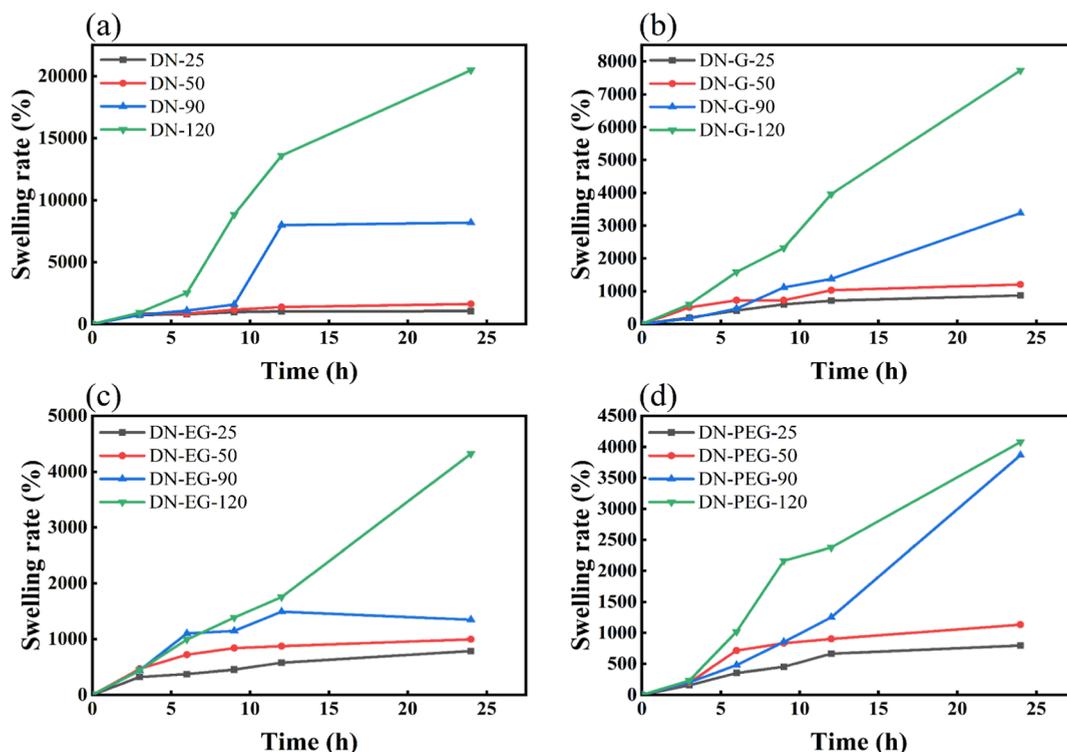
Figure 3. Swelling rate at various temperatures of DN (a) and DN-G (b), DN-EG (c), and DN-PEG (d).
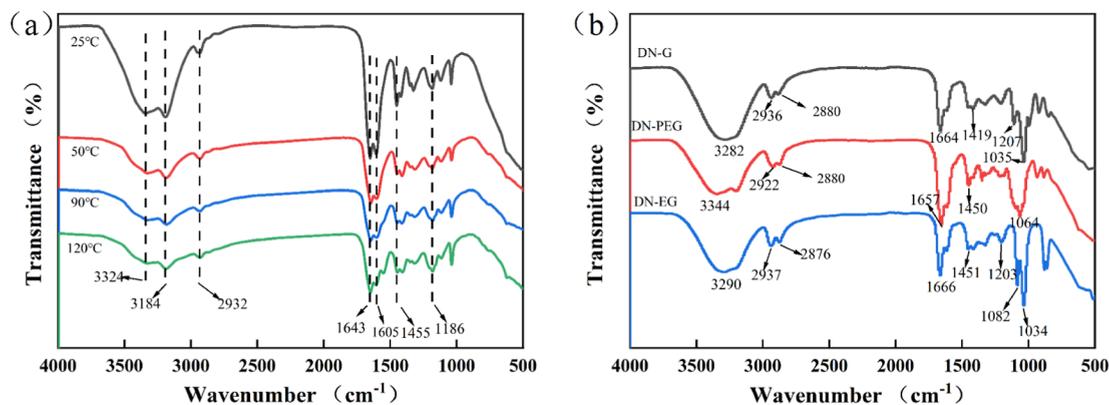
Figure 4. ATR-IR spectra of hydrogels before and after hydrothermal treatment (a) or after immersion in solvents (b).

2.6. PBP Test. The PBP test was applied to test the performance of hydrogel to withstand applied pressures, and the test apparatus is illustrated in Figure 1. PBP is the pressure the hydrogel inside the tapered slot would withhold before further fluid loss was resumed. The fracture mouth width is 5 mm with 0.5 mm width at the end.[27]

The specific experimental steps are as follows: Preparation of water-based mud with formulation as water + 4% bentonite +0.2% Na_2CO_3 + 0.3% xanthan gum. The classic Bingham relationship provides simplified characterization of drilling fluid rheology, and following American Petroleum Institute (API) standards, rheological parameters were measured. The apparent viscosity (AV) was 27.55 mPa·s; the plastic viscosity (PV) was 11.5 mPa·s, and the yield point (YP) was 16.5 mPa·s. The filtrate loss (FL_{API}) of the drilling fluid was 9.0 mL.[28] Hydrogel samples were cut into small pieces in cubic shape and sieved to have their size range from 35 to 45 mm. 10% hydrogel is added to the drilling fluid, and a constant volume

of 2 L slurry is used for testing. The differential pressure is applied to push the hydrogel-laden drilling fluid through the tapered slot and measure the amount of fluid loss. The nitrogen air source pressure is loaded in increments of 0.1 MPa to maximum pressure of 10 MPa, and the pressure at which the plug breaks with obvious fluid loss is noted as PBP.[29]

3. RESULTS AND DISCUSSION

3.1. Structural Characterization of Hydrogels.
3.1.1. Hydrogel Swelling Behavior. As can be seen from Figures 2 and 3, for DN gel, the water content reached 94.16, 99.88, and 99.51% with the swelling rate being 1610, 8183, and 20475% at 50, 90, and 120 °C, respectively. For samples immersed in different solvents, either water content or swelling rate can be reduced at high temperature. At 90 °C, the water content is 97.12, 93.08, and 97.47%, with the swelling rate being 3383, 1346, and 3867% for DN-G, DN-EG, and DN-PEG, respectively. With the temperature being higher, the

difference of water content or swelling rate between DN gel and that after immersion became less, indicating that more solvents will be exchanged by water at high temperature. At 120 °C, the water content is more than 97.5% for DN gel before or after immersion in solvents, and the will gel lose its stability with little strength of gel, as observed in the solution.

According to Paul and John,[30] with temperature increasing, both free energy of polymer mixing in solvent and elastic free energy of the gel network will increase, which can cause more swelling rate and higher water content. Among the three solvent exchange gels, DN-PEG has the lowest water content due to the higher molecular weight of PEG, which can better inhibit water molecules from entering the hydrogel. Solvents in DN gel after immersion can also be exchanged by water in the hydrothermal treatment process, with increasing water content and resulting gel swelling. However, compared to that before immersion, the water content is still less. The polyols contained in EG, PEG, and glycerol react with water molecules to form hydrogen bonds, which reduce the activity of water, thereby reducing the entry of water molecules into the gel with the water content decreasing. PEG contains a large amount of hydroxyl groups, and glycerol, as a polyol, contains three hydroxyl groups, which can react with oxygen ions in water molecules to form hydrogen bonds; they both have better ability to reduce the entry of water molecules into the gel than ethylene glycol, and the water content of DN-PEG and DN-G is lower.

3.1.2. ATR-IR Characterization and SEM Observation. The ATR-IR spectra of the DN gels after hydrothermal treatment at various temperatures are shown in Figure 4a. Peaks at 3324 and 3184 cm^{-1} could be attributed to the stretching vibration of N−H. Characteristic peaks of C=O stretching and N−H deformation were found at 1643, 1605 cm^{-1}, respectively. Alkane −CH$_2$− absorption occurs at 2932 cm^{-1}, and the peak at 1455 cm^{-1} is due to the −CH$_2$− shear vibration. DN gels after aging at 90 °C or below do not show a new absorption peak in comparison, presumably no chemical bond formation, while DN-120 has a new peak at 1550 cm^{-1}. This peak corresponds to the N−O asymmetric stretching. The monomer AM can connect with each other or with the cross-linker BIS to form polyacrylamide gel network by free radical polymerization.[31] As reported by Wellington et al., the fracture mechanism of the PAM chain is caused by the oxidation reaction, which speeds up as the temperature increases, leading to a higher fracture rate of the polymer chain.[32] According to Xiong et al., the cleavage of the cross-linked chain is mainly due to the hydrolysis reaction triggered by the nucleophilic attack of the carbonyl carbon by the water molecule.[31]

Looking at the ATR-IR spectra of DN gels with different solvents (Figure 4b), shifts of characteristic peaks can be observed. For the DN-G sample, 3282 cm^{-1} can be attributed to the OH-stretching peak, and 2936 and 2880 cm^{-1} are for the −CH$_2$− asymmetric and symmetric stretching vibration, respectively. For the DN-PEG sample, 3344 cm^{-1} is the peak for OH-stretching, 2922 and 2880 cm^{-1} are attributed to −CH$_2$− stretching vibration. For the DN-EG sample, 3290 cm^{-1} can be assigned to the OH-stretching, and 2937 and 2876 cm^{-1} are attributed to the −CH$_2$− stretching vibration peaks. C−O stretching occurs at 1110, 1210, and 1203 cm^{-1} for DN-G, DN-PEG, and DN-EG, respectively. Compared to DN gels, C=O stretching peaks shift from 1643 to 1664 cm^{-1} after immersion in glycerol, to 1657 cm^{-1} after immersion in

PEG and to 1666 cm^{-1} after immersion in EG, indicating that hydrogen bonding occurs on the C=O group.[23]

Figure 5a,b shows DN gel after aging at 25 °C (DN-25) or 50 °C (DN-50) as the SEM diagram; it can be clearly seen that
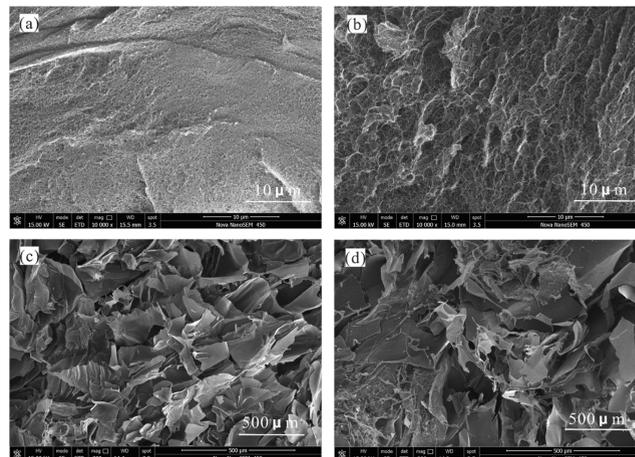
Figure 5. SEM images of DN gel after hydrothermal treatment at 25 (a), 50 (b), 90 (c), and 120 °C (d).

DN-25 has smaller pores than DN-50, which indicates that DN-25 has a tighter gel structure. Figure 5c,d shows SEM diagrams of the gel after aging at 90 °C (DN-90) or 120 °C (DN-120) with much larger size of pores,[2,33] where a small number of breaks can be seen in gel networks for DN-90, and a large number of breaks exists in DN-120, indicating that in the range of 90−120 °C, rupture of cross-linking gel occurs with segments breaking in polymer or cross-linker. The whole possible hydrothermal degradation processes for gel before and after immersion in solvents are proposed in Figure 6.

3.2. Compression Strength and Fracture PBP Test. As can be seen from Figure 7, the gels after immersion in glycerol and PEG200 have a higher compression strength of more than 60 MPa. After hydrothermal treatment at 25 and 50 °C, with increase of water content and swelling rate, large reduction of compression strength was observed for DN gel or the gel after immersion in EG, whose compression strengths are only 3.6 and 0.3 MPa at 50 °C, while for gels after immersion in glycerol and PEG200, compression strength can still reach 55 MPa and 35 MPa, respectively. After hydrothermal treatment at 90 °C, little residual compression strength can be observed for DN gel or DN-EG gel, and compression strength of DN-G gel reduced to 8.4 MPa. Only DN-PEG gel exhibited high compression strength of 16.8 MPa. After aging at 120 °C, only the compression strength of DN-PEG can reach more than 1 MPa, and the compression strength of the remaining three gels was totally lost. The compression strength of DN-EG is the lowest, even lower than DN gel, because EG with lower molecular weight cannot prevent water molecules from entering the gel at high temperatures, resulting in gel swelling and a decrease in cross-linking density.

In addition, among the three solvents, ethylene glycol, as a diol, only contains two hydroxyl groups to react with the groups on the polymer chain, which makes ethylene glycol have the weakest protective effect on the polymer chain. At high temperature, the flexible segment of EG has a detrimental effect on gel, and the compression strength of DN-EG is the weakest. The compression strength of DN-PEG and DN-G is
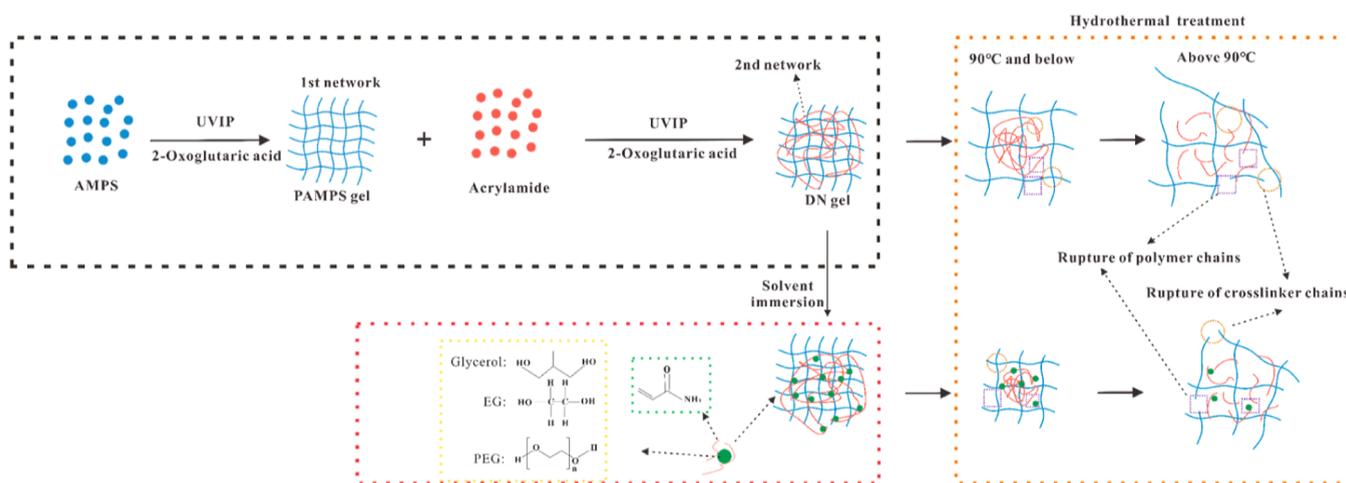
Figure 6. Proposed schematic diagram of gel structure variation during hydrothermal treatment.
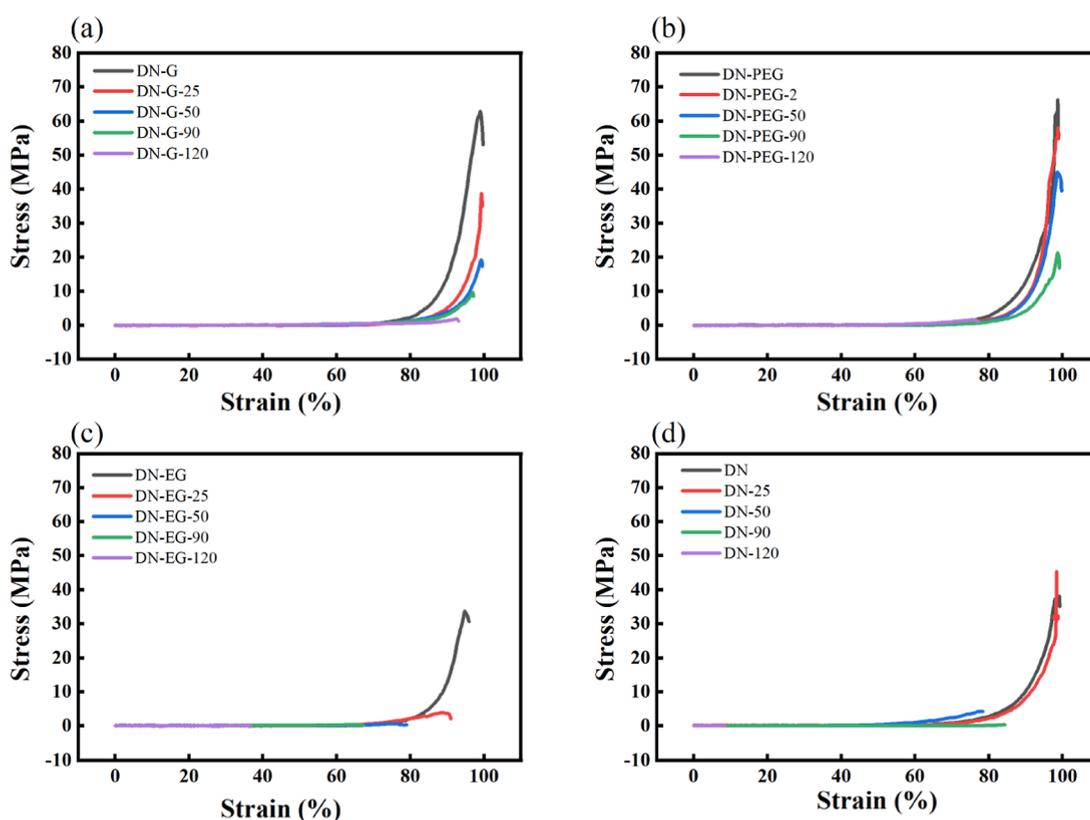
Figure 7. Stress−strain curves of samples DN-G (a), DN-PEG (b), DN-EG (c), and DN (d) before and after hydrothermal treatment at different temperatures.

higher, because PEG and glycerol inhibit the swelling of dual-network hydrogels by displacing water molecules and reducing the activity of water; on the other hand, by reacting with the groups on the polymer chain, the rupture of the polymer chain or cross-linker chains at high temperature is reduced, ensuring the integrity of the hydrogel structure. This gives DN-PEG and DN-G relatively high compression strength. PEG, as a polymer, has a higher molecular weight and more hydroxyl groups, and DN-PEG has a higher compression strength than DN-G.

The compression strength of the gel will directly affect the fracture plugging performance of the gel in the tapered slot, and the stronger the compression strength of the gel, the better

its plugging performance will be. As shown in Figure 8, DN-PEG and DN-G exhibited better plugging performance with higher PBP. After hydrothermal treatments at 90 °C or more, the DN or DN-EG gel was easily ruptured by the applied pressure and total fluid loss occurs at very low pressure. DN-G gel can still withstand about 2 MPa pressure after aging at 90 °C or more, and DN-PEG gel has higher PBP than DN-G.

4. CONCLUSIONS

Dual-network (DN) hydrogels are promising materials for lost circulation treatment due to high strength and toughness, and solvents such as PEG and glycerol can be used to enhance its
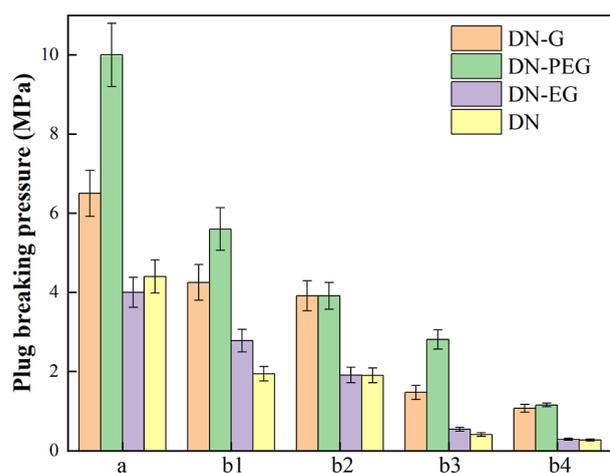
Figure 8. Fracture PBP for gels before hydrothermal treatments (a) or after hydrothermal treatments at 25 (b1), 50 (b2), 90 (b3), and 120 °C (b4), respectively.

strength. The effects of swelling behaviors and solvent exchange on DN gel strength and fracture plugging performance were studied. At high temperature, gel water content and swelling rate will increase with the increase in pore size, which can reduce the cross-linking density of gel, and obvious reduction of compression strength and PBP will occur. After hydrothermal treatment at 90 °C, although no chemical bonding change exists in the gel during swelling, there will be little residual strength for the DN gel. At 120 °C, a new band of N−O asymmetric stretch can be observed, which confirms the rupture of cross-linker chains in the gel. DN gel loses its stability due to rupture of polymer chains and cross-linker chains. After immersion, DN gel strength was greatly improved, and the compression strength can reach 64.5 MPa, with PBP being a maximum of over 10.0 MPa after immersion in PEG. After hydrothermal treatments, part of the solvent molecules can be exchanged by water, and the remaining solvent molecules in the gel can react with water molecules to form hydrogen bonds, which reduces the activity of water, thereby reducing the entry of water molecules into the gel; on the other hand, by reacting with the groups on the polymer chain, the rupture of the polymer chain or cross-linking chain at high temperature is reduced, ensuring the integrity of the hydrogel structure. As a result, the gel after immersion in solvents has a higher compression strength or PBP after hydrothermal treatments. Among the three solvents, PEG with a larger molecular weight and more hydroxyl groups exhibits the best ability. For DN-PEG, compression strength can still reach 16.8 MPa with 2 MPa more of PBP even at 90 °C. However, when the temperature reaches 120 °C, most of the solvents will be exchanged by water molecules and water content is more than 97.5% for DN gel before or after immersion, the effect of solvents was greatly reduced, and the strength of gel was greatly reduced. As a result, under the temperature of 90 °C, DN-PEG was suitable for use in water-based drilling fluid for lost circulation treatments in fractured formation.

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Notes

The authors declare no competing financial interest.

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