Article

Morphology and Rheological Properties of Polyacrylamide/Bentonite Organic Crosslinking Composite Gel

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Abstract: The use of polymer gel for water control and oil addition is a common technical method in oilfield development. The polymer and hydrated bentonite react under the action of an organic crosslinking agent to form a composite gel. The particle-size change and microstructure of the composite gel were analyzed via shear thinning, thixotropic, viscoelastic, and start-up stress rheology experiments. The experimental results show that the polyacrylamide/bentonite organic crosslinked composite gel was a gel system with bentonite as the core aggregate structure, and the large particle-size distribution was mostly increased with increasing crosslinker content. The composite gel presented shear thinning characteristics, the content of bentonite or crosslinking agent was increased, and the shear resistance was stronger at a high shear rate. The composite gel exhibited positive thixotropic properties, and the thixotropy increased with increasing bentonite content. The composite gel had good viscoelastic characteristics, the elastic characteristics of the composite gel showed more significantly with bentonite increases, and the viscosity of the composite gel showed its characteristics more significantly with the crosslinking agent increased. After loading at a rate on the composite gel, the shear stress increased significantly with time and reached its maximum value, and then the shear stress decreased and gradually stabilized.

Keywords: composite gel; aggregate; shear thinning; thixotropic; viscoelasticity; start-up stress experiment

1. Introduction

For the middle and late stages of oilfield development, the water content increases significantly because the original layer, continuous mining, or improper construction operation lead to intensified interlayer heterogeneity and serious turbulence [1–4]. In particular, in the later stage of water injection, large holes with a diameter greater than 30 mm will inevitably appear [5]. As a chemical solution for well effluent oil and gas control, hydrogel is used to treat water intrusion cones in various water sources, reduce the water permeability of heterogeneous reservoirs, and block the channels connecting aquifers [6]. The choice of polymer gel treatment requires consideration of multiple factors, including consistency of treatment issues, application points, state of the gel, stability of the downhole environment, and economic cost [7].

Partially hydrolyzed polyacrylamide (HPAM) is one of the most widely used polymers in oil fields. The polymer solution has a stretched conformation due to the electrostatic repulsion of carboxyl ions on the polymer molecular chain and has a strong viscosity-increasing ability. It has good water solubility and is reasonably priced among many polymer flooding agents [8]. As a thickener and flocculant, it has been widely used in oil exploitation, drilling, mud treatment, and sewage treatment [1]. The mineralization degree of underground mineral deposits is high, the ionic polymer is affected by high concentration of salt ions, and its salt resistance is poor, while the hydrophilic group of polyacrylamide is the non-ionic amino group, which is more sensitive to salt ions. While its molecular chain can form a large number of hydrogen and bond with water, the formed hydrogel has mechanical strength and salt resistance [9,10]. At present, polyacrylamide is mainly used to add a crosslinking agent to form a large pore in the gel sealing oil layer under a certain temperature.

Among many clay minerals, bentonite can be used to prepare nanocomposites because it contains a large amount of MMT [$\text{Na}_{1/3}(\text{Al}_{5/3}\text{Mg}_{1/3})\text{Si}_4\text{O}_{10}(\text{OH})_2$], which consists of aluminum oxyhydroxide octahedral sheets sandwiched between two layers of siloxane tetrahedral sheets [11]. These molecules, which are stacked on top of each other, are held together by the van der Waals force and are separated from each other by a 1 nm slit. These spaces are usually occupied by cations, such as alkali- and alkaline-earth cations (Na and K), which balance the negative charge generated by the isomorphous material in the layer (for bentonite, Al_3 is replaced by Mg_2) [12]. Other cations are replaced, so ordinary bentonite is often sodiumated to form sodium bentonite. Clay is cheap and easy to obtain, and there are many types of it. The most widely studied and widely used is bentonite. The small particles of bentonite swell in water medium to form an inorganic gel material. This composite material can be effectively combined with an organic system through a chemical reaction to compensate for the deficiency of the single material, forming a new development direction, and has certain research and application value in the water-swelling resin material area [13]. In oil field drilling fluid applications, the polymer/clay hybrid nanoparticles significantly improve rheological and filtration properties of the drilling fluids, and they remain stable at high pressure, high temperature, and harsh salinity conditions [14]. In enhanced oil recovery applications, a certain range of clay concentration has positive effects on polymer solution viscosity that should be increased to higher rates to be able to compensate for all negative effects of salinity [15]. In this paper, the composite gel was formed by hydration of bentonite and polyacrylamide under the action of an organic crosslinking agent. The particle size and microstructure of the composite gel were studied, and the internal structure and mechanics of the composite hydrogel were analyzed using rheological methods.

2. Experimental Details

2.1. Raw Materials and Reagents

Polymer: Polyacrylamide (HPAM), molecular weight 15 million (Beijing Hengju Technology Development Co., Ltd.). Crosslinking agent: urotropine (Chengdu Kelong Chemical Reagent Factory), analytical grade; resorcinol (Chengdu Kelong Chemical Reagent Factory), analytically pure; and oxalic acid (Chengdu Kelon Chemical Reagent Factory). Stabilizer: thiourea (Chengdu Kelon Chemical Reagent Factory), analytically pure. Hydrated bentonite system: sodium bentonite (Xin Runda Mining Co., Ltd.); sodium carbonate (Chengdu Kelon Chemical Reagent Factory), chemically pure; and sodium hydroxide (Chengdu Kelon Chemical Reagent Factory), chemically pure. Water: ordinary water.

2.2. Preparation of Polyacrylamide/Bentonite Organic Crosslinked Composite Gel

Sodium bentonite hydration: 0.25% sodium hydroxide and 0.5% sodium carbonate were dissolved in water. Then 15% sodium bentonite was added, and the mixture was stirred well. After standing for 48 h, hydrated bentonite was obtained. Under high-speed agitation, hydrated bentonite, polymer (polyacrylamide), and organic crosslinking agent (urotropine, resorcinol, and oxalic acid) and stabilizer (thiourea) were added in sequence and stirred well. The mixed solution was sealed and placed at 80 °C

for 48 h to obtain a polyacrylamide/bentonite organic crosslinked composite gel (hereinafter referred to as the composite gel). The preparation process is shown in Figure 1. The formulations of the composite gels are shown in Table 1.

Figure 1. Preparation process of polyacrylamide/bentonite organic crosslinked composite gel.

Table 1. Formulations of composite gels.

Sample	Bentonite (mg/L)	HPAM (mg/L)	Urotropine (mg/L)	Resorcinol (mg/L)	Oxalic Acid (mg/L)	Thiourea (mg/L)
BH	40,000	1500	0	0	0	0
BHCT-1	40,000	1500	100	80	100	30
BHCT-2	40,000	1500	200	175	200	30
BHCT-3	40,000	1500	400	350	400	30
BHCT-4	40,000	1500	600	500	600	30
BHCT-5	10,000	1500	200	175	200	30
BHCT-6	20,000	1500	200	175	200	30
BHCT-7	60,000	1500	200	175	200	30
HCT	0	1500	200	175	200	30

2.3. Testing and Characterization

2.3.1. Particle-Size Analysis

The particle-size distribution of the composite gels (BH, BHCT-1, BHCT-2, BHCT-3, and BHCT-4) under different crosslinker contents was analyzed using a Mastersizer 3000 laser-diffraction particle-size analyzer.

2.3.2. Scanning Electron Microscopy Analysis

The microstructure of the composite gel (BHCT-2) was analyzed with the field emission scanning electron microscope (FE-SEM) JSM-7800F after drying on the loading station and compared with the polyacrylamide gel (HCT) microstructure.

2.3.3. Rheology Tests

The advanced rheometer (Anton Paar MCR102) employed in this study uses a cylinder measuring device (rotor type CC27) to measure the composite gel under different bentonite contents (BHCT-5, BHCT-6, BHCT-2, and BHCT-7) and different crosslinking agent contents of the composite gel (BHCT-1, BHCT-2, BHCT-3, and BHCT-4).

1. **Shear Thinning:** The apparent viscosity of the composite gel was determined as a function of shear rate at 80 °C (shear rate from 0.01 to 50 s^{−1}).

2. Thixotropy: At 80 °C, first, the shear rate increased from 0 to 20 s⁻¹ within 50 s. Then, shears at a shear rate of 20 s⁻¹ for 20 s occurred. After that, the shear rate decreased from 20 to 0 s⁻¹ within 50 s. Finally, we measured the area of the thixotropic ring, which consists of the curves of the shear rate and shear stress. The area represents the degree of thixotropic degree of the composite gel.
3. Viscoelasticity: In the oscillation mode, the appropriate strain values for frequency scanning (strain $\gamma = 1\%$, angular frequency $\omega = 0.1\text{--}10$ rad/s), the elastic modulus G' , and loss modulus G'' were tested at 80 °C.
4. Start-up stress test: At 80 °C, the fixed shear rate was 0.3 s⁻¹, and the shear stress was measured instantaneously with time.

3. Experimental Results and Discussion

3.1. Effect of Crosslinker Concentration on Composite Gel Particle Size

The particle-size distribution of the composite gel with different crosslinker contents was analyzed using the Mastersizer 3000 laser-diffraction particle-size analyzer as shown in Figure 2. It can be seen that the particle-size distribution of the composite gel without a crosslinking agent was relatively wide, mainly distributed in the range 100–3000 μm . As the amount of crosslinking agent increased, the particle-size distribution curve shifted to a larger particle size. This is because the degree of crosslinking of polyacrylamide increased. The crosslinked polyacrylamide was easily encapsulated with hydrated bentonite to form large aggregates; hence, the proportion of coarse particles increased, as shown in Figure 3.

Figure 2. Particle-size distribution of composite gel under different crosslinker concentrations.

Figure 3. Schematic of particle-size change of composite gel under different crosslinker concentrations.

The particle-size distribution of composite gel under different crosslinker concentrations is shown in Table 2. From the particle-size statistics, the particle cumulative distribution of 10% Dv(10) gradually became smaller as the concentration of the crosslinking agent increased. The value of the particle cumulative distribution of 50% Dv(50) and 90% Dv(90) became larger, which indicates that increasingly more polyacrylamide and bentonite adsorbed and crosslinked to form aggregates with larger molecular volume, and so the particle size was larger. The composite gel is a gel system with bentonite as the core aggregate.

Table 2. Particle-size distribution of composite gel under different crosslinker concentrations.

Sample	Specific Surface Area (m ² /kg)	Dv(10) (μm)	Dv(50) (μm)	Dv(90) (μm)
BH	375.6	10.7	50.6	203
BHCT-1	407.3	8.06	73.4	724
BHCT-2	540.7	4.27	54.4	868
BHCT-3	770.3	2.84	89.2	1850
BHCT-4	807.8	2.51	950	2460

3.2. Microstructure of Polyacrylamide/Bentonite Organic Crosslinked Composite Gel

The microstructure of the composite gel (BHCT-2) and the polyacrylamide gel (HCT) is shown in Figures 4 and 5. Comparing the microstructures of the two samples, it can be seen that the polyacrylamide gel (HCT) exhibited a porous medium form, and the inside structure was mainly composed of polymer aggregate formed by intermolecular crosslinking. Different from the polyacrylamide gel (HCT), the structure of the composite gel (BHCT-2) exhibited a sheet-like microstructure and was characterized by the structure of bentonite. This shows that the composite gel was an aggregate structure with bentonite as the core, and the crosslinked polymer was adsorbed on the surface of the bentonite.

Figure 4. Composite gel (BHCT-2) microstructure.

Figure 5. Polyacrylamide gel (HCT) microstructure.

3.3. Rheological Properties of Composite Gel

3.3.1. Test of the Flow Curve of Composite Gel

The viscosity change of the composite gel under different bentonite concentrations is shown in Figure 6. It can be seen from the figure that the composite gel exhibited shear thinning characteristics, and the viscosity of the composite gel decreased with increasing shear rate. The bentonite of the composite gel showed different degrees of orientation in the flow direction, and polyacrylamide also exhibited different degrees of stretching, as shown in Figure 7. Bentonite is a flaky particle with a negative charge on the plane and a positive charge on the end face. In the bentonite–water suspension, bentonites have face-to-face (FF), edge-to-edge (EE), and face-to-edge (FE) connection methods. The FF mode can lead to thickening and increase of clay particles, while EE and FE connections can easily form a three-dimensional network structure of a “house of cards” between bentonite particles [11,12]. The structural viscosity of the aggregate formed in the solution was low when the amount of bentonite was relatively small, and the aggregate formed with the crosslinked polyacrylamide was less. When the content of bentonite increased, more aggregates were formed, and the distance between the aggregates became smaller, and the interaction increased, forming a strong spatial network structure, especially at low shear rates. The “house of cards” was formed by the mutual attraction of the aggregates and had a strong three-dimensional network structure and a high viscosity of the composite gel. From the viscosity data under high-shear-rate conditions, it can be seen that the viscosity of composite gel increased with increasing bentonite content. The kinetic volume of water was relatively large, indicating that the bentonite in the gel had a stable structure and good shear resistance under high-shear-rate conditions.

Figure 6. Viscosity curve of composite gels at different bentonite concentrations.

Figure 7. Internal structure changes under shear flow of composite gel.

The viscosity change of the composite gel under different crosslinker concentrations is shown in Figure 8. It can be seen from the figure that the composite gel formed by different concentrations of the crosslinking agent still exhibited shear thinning characteristics after shearing. The viscosity of the composite gel formed by the high-concentration crosslinking agent is relatively lower than that of the low-concentration crosslinking agent. This is because the structure of the composite gel formed by polyacrylamide adsorbed on the bentonites at a low concentration of the crosslinking agent. The polyacrylamide was partially adsorbed on bentonite, and the rest can be extended in water. Therefore, the viscosity of the gel was formed by the structure of bentonites and the structure of entangled polyacrylamide in aqueous solution. Under these conditions, the composite gel had a higher viscosity at a low shear rate, and the shear thinning performance was more obvious at a high shear rate. When the crosslinking agent concentration was increased, the polyacrylamide crosslinked and formed aggregate with bentonites. The structures of the composite gel were gradually transformed into bentonite as the structure mainly formed, and the structure formed by the intercalation of polyacrylamide in aqueous solution was decreased. Owing to the reduced structure of polyacrylamide stretching in aqueous solution and the volume of aggregate with bentonite becoming large after crosslinking, the viscosity of the composite gel decreased at a low shear rate and increased at a high shear rate.

Figure 8. Viscosity curve of composite gel with different crosslinker concentrations.

3.3.2. Test of Thixotropy of Composite Gel

Thixotropy is an important characteristic in rheology. It refers to a rheological phenomenon in which the viscosity or shearing force of a system changes with time under agitation or other mechanical action. The characteristics have important guiding significance for the structural strength and structural change of a material. The area of the thixotropic ring can qualitatively indicate the relative size of the thixotropy of the sample, and a larger area of the thixotropic ring indicates that the system has strong

thixotropy. When shearing is applied, if the rate of destruction of the internal structure is greater than the rate of reconstruction, the system will exhibit positive thixotropic properties, and vice versa, i.e., negative thixotropy [16].

The effect of bentonite addition on the thixotropy of the composite gel is shown in Figure 9, and the area of each thixotropic ring is calculated in Table 3. The experimental results show that the composite gel had a thixotropic ring, the upper line of which was larger than the lower line and exhibited positive thixotropic characteristics. Bentonite had a great influence on the thixotropic properties of the composite gel. With increasing bentonite content, the area of the thixotropic ring increased significantly, and the thixotropy increased obviously, indicating that bentonite could greatly increase the thixotropic structure of the composite gel and required a longer time to rebuild the internal structure.

Figure 9. Shear thixotropy of composite gels at different bentonite concentrations.

Table 3. Thixotropic ring area of composite gels of different bentonite concentrations.

Sample	BHCT-5	BHCT-6	BHCT-2	BHCT-7
Thixotropic ring area	0.224 Pa/s	2.785 Pa/s	13.482 Pa/s	38.597 Pa/s

The effect of crosslinking agent addition on the thixotropy of the composite gel is shown in Figure 10. The area of each thixotropic ring is calculated in Table 4. The experimental results show that the composite gel had a large thixotropic ring area and strong thixotropy under the condition of low crosslinking agent content. With increasing crosslinking agent, the area of the thixotropic ring of the composite gel decreased and the thixotropy decreased. Under a low concentration of crosslinking agent, the three-dimensional structure of composite gel formed by bentonite flaky particles and the polyacrylamide entanglement was strong, and the rate lag was obvious. The shear field destroyed its structure and required more energy, and it took longer to recover after the structure was destroyed. When the concentration of the crosslinking agent was increased, the crosslinking density of the composite gel increased, the thickening property based on the formation of aggregates with

bentonite as the core was gradually formed, and the polyacrylamide molecular chain entanglement between the aggregates was greatly reduced. The rate of lag was reduced, the speed of the recovery structure increased, and the thixotropic area decreased.

Figure 10. Shear thixotropy of composite gels at different crosslinker concentrations.

Table 4. Area of thixotropic ring of composite gel under different crosslinker concentrations.

Sample	BHCT-1	BHCT-2	BHCT-3	BHCT-4
Thixotropic ring area	10.032 Pa/s	13.482 Pa/s	4.926 Pa/s	4.741 Pa/s

3.3.3. Test of Viscoelasticity of Composite Gel

The elastic modulus and loss modulus of different contents of bentonite composite gels as a function of shear angle frequency are shown in Figures 11 and 12, respectively. It can be seen from the figures that the elastic modulus G' and loss modulus G'' were significantly improved with an increasing amount of bentonite when the bentonite content reached 4%. The structural strength increased due to the enhanced network structure formed by the layered structure of bentonite. With increasing frequency, the elastic modulus and loss modulus increased greatly, and the large-sized aggregates were transformed into elastic storage due to the deformation under the external force; hence, the higher the frequency, the better the elasticity. When the bentonite content increased, the elastic modulus and loss modulus of the composite gel were increased because the number of aggregates formed by crosslinked polyacrylamide and bentonite increased, and the distance between aggregates became smaller. The spatial structure of the composite gel was denser, and the three-dimensional network structure of the “house of cards” formed was strong.

Figure 11. Elastic modulus of composite gels with different bentonite contents.

Figure 12. Loss modulus of composite gels with different bentonite contents.

The trend of $\tan\delta$ is shown in Figure 13. As the amount of bentonite increased, the $\tan\delta$ value decreased. Because $\tan\delta$ is the ratio of the loss modulus to the elastic modulus, the smaller the $\tan\delta$, the larger elasticity of the sample, indicating that the network structure is more complete. The larger the $\tan\delta$, the less the network formed by the sample is in a viscous flow state, and the viscosity ratio becomes larger [16]. As the amount of bentonite increased, the $\tan\delta$ value of the composite gel decreased. From the rheological point of view, the composite gel of crosslinked bentonite and polyacrylamide formed a strong physical network, the elastic property was increased with the addition of more bentonite, and the network structure was more complete.

Figure 13. Loss factor of composite gels with different bentonite contents.

Next, the amount of bentonite and polymer was kept unchanged, and the content of the crosslinking agent was changed. The elastic modulus and loss modulus of the composite gel as a function of the shear angle frequency are shown in Figures 14 and 15, respectively. The elastic modulus of the composite gel decreased with increasing crosslinker agent content, and the loss modulus increased compared with the addition of bentonite. The viscosity of the gel was formed by the structure of bentonite and the structure of entangled polyacrylamide in aqueous solution, and the crosslinking of polyacrylamide and the formation of aggregates with bentonite increased (Figure 2 experimental results) with increasing degrees of crosslinking. The aggregate of bentonite became the main structure for composite gels, and the ability of polyacrylamide to entangle each other in aqueous solution was weakened, resulting in weaker elastic deformation between gels; hence, solid-state properties of composite gels were weakened, and the elastic modulus decreased. The loss modulus increased because the hydrodynamic volume of the aggregate formed by crosslinking increased. When the crosslinking agent content was further increased, the aggregate formed by crosslinking further increased, and there was a certain hydrogen bond attraction between the aggregates. The solid-state characteristics were enhanced, and the elastic modulus increased slightly.

Figure 14. Elastic modulus of composite gel with different crosslinker contents.

Figure 15. Loss modulus of composite gel with different crosslinker contents.

The trend of $\tan\delta$ is shown in Figure 16. As the amount of crosslinking agent increased, the $\tan\delta$ value increased, indicating that the proportion of the viscous characteristics of the composite gel became larger, and the elastic characteristics were lowered. The result shows that the structure formed by aggregates increased, the hydrodynamic volume increased, and the network of sample formation was incomplete.

Figure 16. Loss factor of composite gel with different crosslinker contents.

3.3.4. Test of Start-up Stress of Composite Gel

The start-up stress test is a time-dependent apparent shear stress of the gel used to directly characterize the shear stress required for internal structural damage [16]. Figure 17 shows the time-dependent apparent shear stress of the composite gel with different bentonite contents, and Table 5 shows the maximum value of the apparent shear stress. The results show that when the shear rate began to be loaded onto the composite gel, the apparent shear stress increased significantly with time and reached the highest value. The maximum apparent shear stresses at 1 s were 0.760 Pa (for BHCT-2, with a bentonite content of 40,000 mg/L) and 2.837 Pa (for BHCT-7, with a bentonite content of 60,000 mg/L). When the bentonite content was low, the time required for the apparent shear stress to reach the maximum was longer, and the maximum apparent shear stress value decreased. For a decreasing distance between the composite gel aggregates formed by the increase of bentonite, the

aggregates were more likely to contact each other to form a stronger internal structure after the shear rate was loaded; hence, the shear stress increased. After the shear stress reached a certain value, the internal structure of the composite gel began to break, and the shear stress decreased. The aggregate had different degrees of orientation in the flow direction. At this time, the composite gel gradually showed shear flow, and the shear stress also tended to be stable.

Figure 17. Time-dependent apparent shear stress of composite gels under different bentonite contents.

Table 5. Maximum shear stress of composite gel under different bentonite content.

Sample	BHCT-5	BHCT-6	BHCT-2	BHCT-7
Maximum shear stress	0.090 Pa	0.299 Pa	0.760 Pa	2.837 Pa

The apparent shear stress of the time-dependent composite gel content at different crosslinking agent contents is shown in Figure 18. Table 6 shows the maximum value of the apparent shear stress. When the shear rate began to load onto the composite gel, the time-dependent apparent shear stress curve showed a similar trend as the experimental results in Figure 17. However, the apparent shear stress maximum with increasing crosslinking agent content first decreased and then increased. The structure of the intertwining and the contact of the bentonite was strong when the crosslinking agent had a low concentration because the composite gel exhibited that the polyacrylamide portion was adsorbed on the bentonite, and both were entangled with each other. After the crosslinking agent content further increased, the composite gel was transformed into an internal structure mainly composed of bentonite as the core aggregate. The initial volume of the aggregate was small, and the internal structure of the composite gel was weak; hence, the maximum shear stress required for destroying the internal structure was lowered. However, as the crosslinking agent was further increased, the aggregate was further enlarged, the interaction between the aggregates was enhanced, and the maximum shear stress required to destroy the internal structure gradually increased.

Table 6. Area of thixotropic ring of composite gel under different crosslinking agent conditions.

Sample	BHCT-1	BHCT-2	BHCT-3	BHCT-4
Maximum shear stress	1.146 Pa	0.760 Pa	1.147 Pa	1.450 Pa

Figure 18. Time-dependent apparent shear stress of composite gels under different crosslinker contents.

4. Conclusions

- The composite gel formed by polyacrylamide and bentonite and organic crosslinking agent and stabilizer under high-temperature conditions was formed by polymer adsorption in bentonite and intermolecular crosslinking. The aggregate structure with bentonite as the core was different from the polyacrylamide gel structure formed by polymer aggregates. A larger aggregate particle size was formed with a higher content of crosslinking agent.
- The composite gel exhibited shear thinning characteristics, the bentonite content increased, and more aggregates were formed. The distance between the aggregates became smaller, and the interaction increased and formed a strong spatial network structure. The composite gel had a higher viscosity under shear and shear resistance at a high shear rate. As concentration of the crosslinker increased, the aggregate formed by the composite gel became larger, and the hydrodynamic volume increased. Shear resistance ability was also enhanced.
- The composite gel exhibited positive thixotropic properties and good viscoelastic properties. The area of the thixotropic ring was significantly increased, and the thixotropy was obviously enhanced. When the content of the bentonite increased, the elastic modulus and loss modulus were increased. The elastic performance was increased more, and the network structure was more complete. When the crosslinking agent content increased, the aggregates gradually became larger, and the molecular chain entanglement between the aggregates was greatly reduced. The thixotropy was weakened, and the elastic modulus decreased. The loss modulus increased, and the viscosity characteristics were enhanced, owing to the hydrodynamic volume increase.
- When the shear rate began to be loaded onto the composite gel, the apparent shear stress increased significantly with time and reached the highest value. Then the internal structure of the composite gel began to break, and the shear stress decreased. The flow direction of the aggregate showed different degrees of orientation. At this time, the composite gel gradually showed shear flow, and the shear stress tended to be stable. Start-up stress was apparently improved with the content of bentonite increased. The maximum value of start-up stress first decreased and then increased with increasing crosslinking agent content. The time of the start-up stress became short.

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Abbreviations

γ	strain
ω	angular frequency, rad/s
G'	elastic modulus, Pa
G''	loss modulus, Pa
$Dv(10)$	the particle cumulative distribution is 10%, μm
$Dv(50)$	the particle cumulative distribution is 50%, μm
$Dv(90)$	the particle cumulative distribution is 90%, μm
$\tan\delta$	Loss factor

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