

RESEARCH ARTICLE

High-performance water-based drilling fluid system based on environmentally friendly polymers

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In the exploration and development process of oil and gas resources, drilling operations are a key link in obtaining underground resources. The performance of drilling fluid directly affects many important aspects such as drilling efficiency, wellbore stability, and oil and gas reservoir protection, and more stringent requirements have been put forward for the performance of drilling fluid systems. Meanwhile, with the increasing awareness of environmental protection and the deepening of green chemistry concepts, the development of environmentally friendly high-performance water-based drilling fluid systems has become a research hotspot in the petroleum industry. However, current research has shortcomings such as insufficient temperature and salt resistance, and poor control of filtration losses. This study built an environmentally friendly high-performance water-based drilling fluid system and designed and optimized the filtrate reducer. In addition, the testing indicators and characterization methods of the materials were also investigated and optimized. The results showed that the optimal reaction temperature, pH, and monomer molar ratio for the preliminary polymer filtrate reducer were 62°C, 8.0, and 7:2:1, respectively. The optimal dosage of modified montmorillonite nanomaterials for the final polymer filtrate reducer was 1.1 wt%. In the polymer decomposition, the mass losses of the preliminary and final polymer filtrate reducers prepared at a temperature of 300°C were 12.57% and 19.58%, respectively. The apparent viscosities of the final prepared polymer filtrate reducer, carboxymethyl cellulose, and sulfonated melamine formaldehyde resin were 20.5 mPa·s, 13.75 mPa·s, and 15.87 mPa·s, respectively. The amount of filtrate reducer added was 2.4%, the total amount of sulfonated phenolic resin and sulfonated brown coal resin was 5.0%, the amount of two plugging agents added was 3.0% and 2.0%, and the amount of lubricant added was 1.2%. The designed filtrate reducer and water-based drilling fluid system had good performance, which could ensure the smooth progress of oil drilling operations and stabilize the wellbore. This research provided a new high-performance and environmentally friendly water-based drilling fluid system, which exhibited more efficient and environmentally friendly solutions for drilling operations under complex geological conditions, and promoted the green and sustainable development of drilling fluid technology.

Keywords: polymer; drilling fluid; filtrate reducers; montmorillonite; filtration loss.

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Introduction

In the exploration and development process of oil and gas resources, drilling operations are a key link in obtaining underground resources. As the

"blood" of drilling engineering, the performance of drilling fluid directly affects many important aspects such as drilling efficiency, wellbore stability, and oil and gas reservoir protection [1, 2]. With the continuous growth of global energy

demand, the scale and difficulty of drilling operations continue to increase, and more stringent requirements have been put forward for the performance of Drilling Fluid Systems (DFSs). Meanwhile, influenced by environmental awareness and the concept of green chemistry, environmentally friendly high-performance water-based DFSs have become a research hotspot in the petroleum industry [3, 4]. The commonly used methods for addressing these issues include the selection and formulation of treatment agents, performance evaluation and regulation, and on-site application and improvement. The selection and formulation of treatment agents involves filtrate reducers, thickening agents, lubricants, etc. [5, 6]. Meanwhile, the filtrate reducers have been optimized by many scientists.

Huang *et al.* adopted three types of filtrate reducers including potassium humate, hydroxypropyl starch, and hydrolyzed polyacrylonitrile sodium salt to analyze the effect of filtrate reducers on hydrate formation. The research was conducted at an initial pressure of 6 megapascals, and the results showed that, at 4°C, higher initial pressure resulted in earlier and faster generation of methane hydrates in the filtrate reducer solution [7]. Chang *et al.* synthesized a novel zwitterionic quaternary copolymer in aqueous solution through free radical copolymerization reaction to optimize the water-based drilling fluids. The structure was characterized by Fourier Transform Infrared Spectroscopy (FTIS) and nuclear magnetic resonance. The medium pressure filtration losses of the material before and after hot rolling at 260°C for 16 h were 3.4 mL and 6.0 mL, respectively [8]. Balaga *et al.* explored the thermal degradation of various acrylamide copolymers in different monovalent salt brines to maintain the performance of water-based drilling mud. After aging at 121°C for 16 hours, the high-temperature and high-pressure (HTHP) filtration loss of these copolymers was 18 mL, which provided a valuable tool for drilling automation [9]. However, current research has certain shortcomings such as insufficient temperature

and salt resistance and poor control of filtration loss.

To build a high-performance water-based DFS and improve the performance of the filtrate reducer, this study selected various materials such as acrylamide, anhydrous ethanol and montmorillonite as raw materials and proposed an environmentally friendly aqueous solution polymerization method. The materials in the water-based DFS were also optimized. The research aimed to design a water-based DFS with excellent comprehensive performance, which could meet drilling needs under complex geological conditions while minimizing the impact on the environment. This research introduced improved montmorillonite nanomaterials, which had improved the performance of the filtrate reducer. The study not only provided more efficient and environmentally friendly solutions for drilling operations under complex geological conditions in the petroleum industry but also promoted the green and sustainable development of drilling fluid technology, providing new ideas and methods for materials science and application research in related fields.

Materials and methods

Preparation of polymer filtrate reducer

To optimize the filtrate reducer, different raw materials including acrylamide (Kelon Chemical Reagent Factory, Chengdu, Sichuan, China), montmorillonite (Thermo Fisher Scientific, Shanghai, China), hexadecyltrimethylammonium bromide (McLean Biochemical Technology Co., Ltd., Shanghai, China), N-acryloyl morpholine (Sigma Aldrich, Shanghai, China), ammonium persulfate (Sigma Aldrich, Shanghai, China), sodium p-styrenesulfonate (McLean Biochemical Technology Co., Ltd., Shanghai, China), acryloyl morpholine (Beihua Fine Chemical Co., Ltd., Beijing, China), sodium bisulfite (Sigma Aldrich, Shanghai, China), and anhydrous ethanol (Kelon Chemical Reagent Factory, Chengdu, Sichuan, China) were selected for this study. The

preparation of polymer filtrate reducer included preliminary preparation, preparation of modified nanomaterials, and combination of preliminary prepared materials and modified nanomaterials. In the preliminary preparation of polymer filtrate reducer, aqueous solution polymerization method was employed, which used water as a solvent to dissolve monomers and underwent polymerization reaction under the action of an initiator to generate polymers [10-12]. The advantages of this method were environmental friendliness, safety, low cost, good product performance, and easy operation. Briefly, sodium p-styrenesulfonate, deionized water, acrylamide, and acryloyl morpholine were mixed until all monomers were dissolved. The solution pH was adjusted using sodium hydroxide to 7 - 8. Meanwhile, nitrogen gas was introduced into the solution for half an hour, while deionized water containing redox initiators was added to the solution after the first nitrogen gas injection followed by a second nitrogen gas injection. The crude product was obtained through a water bath and washed before dried and crushed using a WFS-200 Universal Crusher (Zhongmian Machinery Equipment Co., Ltd., Beijing, China). The modified nanomaterials were prepared by using montmorillonite to prepare soil slurry followed by curing for one day. 400 mL of soil slurry was placed in a three necked flask and incubated in a digital constant temperature water bath and stirred for half an hour. Meanwhile, the hexadecyltrimethylammonium bromide was added to deionized water and placed in a flask, while nitrogen gas was introduced into the flask. The reaction was incubated at 80°C for 10 hours before stratifying the suspension and obtaining the bottom solution. The sample was washed and purified multiple times before drying and crushing and screening to obtain the final modified montmorillonite nanomaterials. The final polymer filtrate reducer was obtained by combining the initially prepared polymer filtrate reducer with modified montmorillonite nanomaterials using *in-situ* intercalation polymerization method, a widely used technique in the field of polymer material preparation,

which played a unique role in the preparation of polymer/layered inorganic nanocomposites and could enhance material properties, achieve molecular level composite, and improve material processing performance [13-15]. Briefly, the final polymer filtrate reducer was placed in deionized water and mixed with modified montmorillonite nanomaterials. Sodium styrene sulfonate, acrylamide, and acryloyl morpholine were then added to the mixture and stirred for half an hour before adjusting the solution pH to 8 using sodium hydroxide. Meanwhile, nitrogen gas was passed through the solution for half an hour. After the first nitrogen injection, ammonium persulfate and sodium bisulfite as the initiators were added to the solution followed by a second nitrogen injection and placed in a water bath for 6 hours. The sample was washed and purified multiple times before being dried and crushed to obtain the final polymer filtrate reducer.

Testing and characterization methods for materials

To analyze the performance of the final polymer filtrate reducer, X-ray diffraction results obtained using X'Pert³ Powder X-ray diffractometer (Malvern Panaco, Shanghai, China), FTIS results obtained using GD26-FTIR-650 Fourier transform infrared spectrometer (Dens Instrument Technology, Shanghai, China), thermogravimetric analysis results using Pyris 1 TGA thermogravimetric analyzer (Perkin Elmer, Shanghai, China), material dosage, temperature resistance, salt resistance, filter loss reduction, and lubricity were selected as indicators [16, 17]. Further, the optimal synthesis conditions for the preliminary preparation of polymer filtrate reducers involving reaction temperature, reaction acidity and alkalinity, and monomer molar ratio were investigated, while the dosage of modified montmorillonite nanomaterials in the final polymer filtrate reducer was analyzed. For the construction of the DFS, the indicators of apparent viscosity, plastic viscosity, dynamic shear force, medium pressure filtration loss, and HTHP filtration loss were used for optimal selection [18, 19]. The heating rate was set as 10°C/min with the ambient temperature being

maintained at 800°C. The concentrations of the final polymer filtrate reducer were tested as 0%, 0.6%, 1.2%, 1.8%, 2.4%, and 3.0%, respectively, while the temperatures of 150, 170, 190, 210, and 230°C were also tested. The different sodium chloride concentrations of 0%, 2%, 4%, 6%, 8%, and 10% were further examined. The lubricity was determined by setting the pressure at 180 MPa and a speed of 80 rpm using OFI 112-00-1-C EP extreme pressure lubricator (OFI Testing Equipment, Inc., Houston, TX, USA). The reduction rate (X) of lubrication coefficient was determined as follows [20].

$$X = \frac{X_{ori} - X_{add}}{X_{ori}} \times 100\% \quad (1)$$

where X_{ori} was the lubrication coefficient of the base slurry. X_{add} was the lubrication coefficient after introducing the treatment agent. In terms of apparent viscosity and plastic viscosity, a six-speed viscometer was used to measure the viscosities at different rotational speeds. Dynamic shear force was the difference between apparent viscosity and plastic viscosity. The medium pressure filtration loss was achieved through a medium pressure water loss test with the pressure being set at 0.70 MPa. The HTHP filtration loss was achieved through HTHP water loss testing, and the numerical measurement was required after meeting the set temperature for half an hour.

Construction of drilling fluid system

This study incorporated the final polymer filtrate reducer into the drilling fluid formula by selecting the polysulfide drilling fluid formulation as the basis for the DFS. The key components of sulfonated material ratio, lubricant, and plugging agent were optimized. In addition, the formula also included some basic components including 400 mL of water, 4.0% bentonite, 0.5% sodium hydroxide, 20% sodium chloride, and 200 g of barite. Eventually, both the sulfonated material ratio (S) and the lubricant containing six different schemes (L) were tested. The sulfonated material ratio of $S1$ to $S6$ corresponded sulfonated

phenolic resins of 3.0%, 5.0%, 5.0%, 4.0%, 7.0%, and 6.0%, respectively, while their corresponding sulfonated ligand resins were 5.0%, 3.0%, 5.0%, 7.0%, 4.0%, and 6.0%, respectively. For the lubricant $L1$ to $L6$, their corresponding Type 1 values were 3.0%, 0%, 0%, 3.0%, 2.0%, and 2.5%, respectively, while their corresponding Type 2 values were 0%, 3.0%, 4.0%, 2.0%, 3.0%, and 2.5%, respectively. For the plugging agent, five schemes were examined as 0.3%, 0.6%, 1.2%, 1.6%, 2.2% and named as $P1$ to $P5$.

Results and discussion

Verification of optimal synthesis conditions for polymers

A comparative verification was conducted to clarify the optimal synthesis conditions for the preliminary and the final polymer filtrate reducers. The effects of different reaction temperatures and acidity/alkalinity on the material in the preliminary polymer filtrate reducers showed that, as the reaction temperature increased, the apparent viscosity of the initially prepared polymer filtrate reducer first increased and then decreased, while the medium pressure filtration loss and HTHP filtration loss both decreased first and then increased. The temperature corresponding to the trend changes of the three indicators was 62°C, and the respective values of indicators were 25.3 mPa·s, 6.78 mL, and 24.43 mL (Figure 1a). Under the reaction pH, the changes in apparent viscosity, medium pressure filtration loss, and HTHP filtration loss were consistent with those at the reaction temperature, and the pH at which the trend turning point was 8. At this time, the values of the three indicators were 29.5 mPa·s, 5.58 mL, and 18.45 mL, respectively (Figure 1b). Therefore, the optimal reaction temperature and pH for the preliminary polymer filtrate reducer were 62°C and 8, respectively. The influence of monomer ratio and modified montmorillonite nanomaterials on polymers demonstrated that the maximum values of apparent viscosity, medium pressure filtration loss, and HTHP filtration loss were 30 mPa·s, 13.5 mL, and 43.3

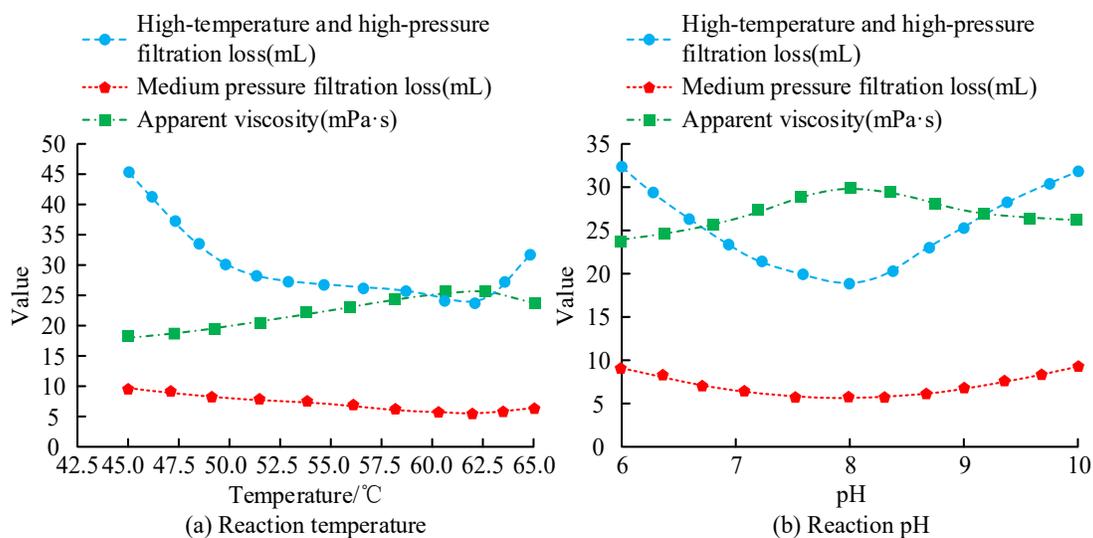


Figure 1. The influence of different reaction temperature and reaction pH on materials.

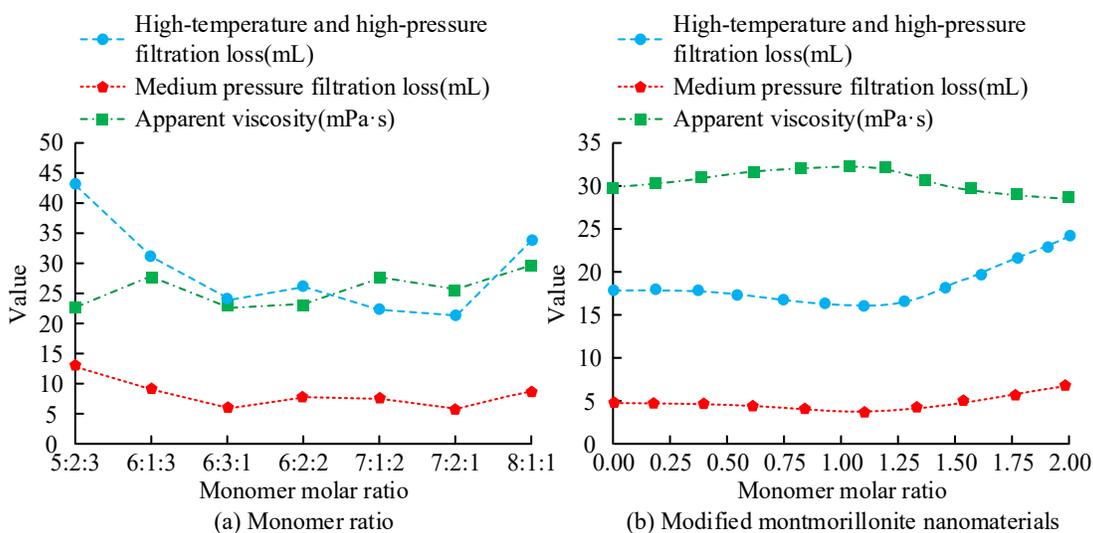


Figure 2. The influence of monomer ratio and modified montmorillonite nanomaterials on polymers.

mL, respectively, while their corresponding monomer molar ratios were 8:1:1, 5:2:3, and 5:2:3, respectively. Meanwhile, the minimum values corresponding to these three indicators were 23 mPa·s, 6.37 mL, and 21.9 mL with their corresponding monomer molar ratios of 5:2:3, 7:2:1, and 7:2:1, respectively (Figure 2a). With the increase of the modified montmorillonite nanomaterials, the apparent viscosity of the final polymer filtrate reducer first increased and then decreased, while the medium pressure filtration

loss and HTHP filtration loss both decreased first and then increased. In addition, the amount of modified montmorillonite nanomaterials corresponding to the trend changes of the three indicators was 1.1 wt% (Figure 2b). Therefore, the optimal monomer molar ratio for the preliminary polymer filtrate reducer was 7:2:1, and the optimal modified montmorillonite nanomaterials for the final polymer filtrate reducer was 1.1 wt%.

Characterization of polymer materials

X-ray diffraction, FTIS, and thermogravimetric analysis were used to characterize the final preparation of polymer filtrate reducer. Preliminary preparation of polymer filtrate reducer and modified montmorillonite nanomaterials were also used for comparison. The X-ray diffraction analysis of different materials demonstrated that, within 6° - 8° , the preliminary and final prepared polymer filtrate reducers did not have clear diffraction peaks, while modified montmorillonite nanomaterials exhibited clear diffraction peaks (Figure 3). This result might be due to the *in-situ* polymerization of monomers between the layers of montmorillonite, resulting in excessive interlayer spacing of montmorillonite, which made it impossible to display diffraction peaks in X-ray diffraction analysis.

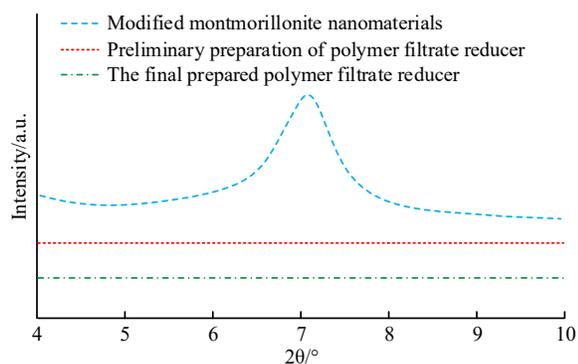


Figure 3. X-ray diffraction analysis of different materials.

The FTIS analysis of different materials showed that both the preliminary and final prepared polymer filtrate reducers contained numerous characteristic peaks, and all characteristic peaks of the preliminary prepared polymer filtrate reducer could be found in the final prepared polymer filtrate reducer. In addition, the final prepared polymer filtrate reducer still exhibited characteristic peaks at $525.0/\text{cm}$ and $796.0/\text{cm}$ (Figure 4a). In the modified montmorillonite nanomaterials, these two characteristic peaks could be found to correspond to each other (Figure 4b). The modified montmorillonite

nanomaterial layer had been successfully added to the polymer.

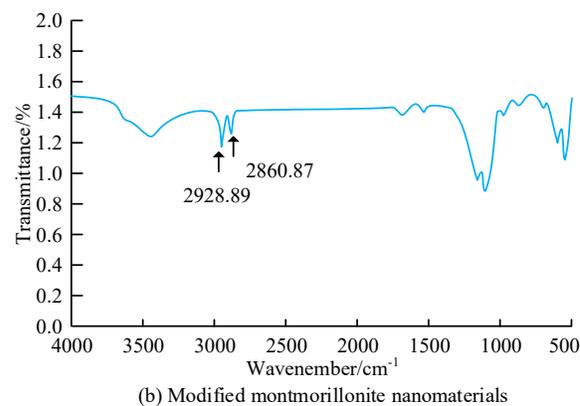
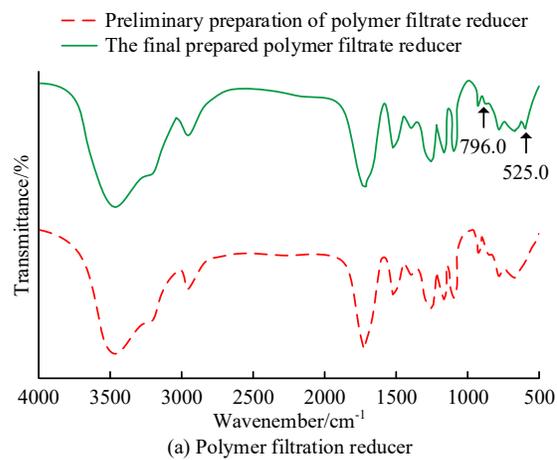


Figure 4. Fourier transform infrared spectroscopy analysis of different materials.

The thermogravimetric analysis of different materials showed that the weight loss of montmorillonite before modification was significantly greater than that after modification within $50 - 240^{\circ}\text{C}$. At 210°C , the total weight loss of montmorillonite material before and after modification was 7.34% and 3.13%, respectively (Figure 5a). The modified montmorillonite nanomaterials had stronger temperature resistance. At 240°C , the total weight loss of the final prepared polymer filtrate reducer exceeded that of the initially prepared polymer filtrate reducer. However, when the temperature exceeded 240°C , the situation reversed. At 300°C , the mass loss of the preliminary and final

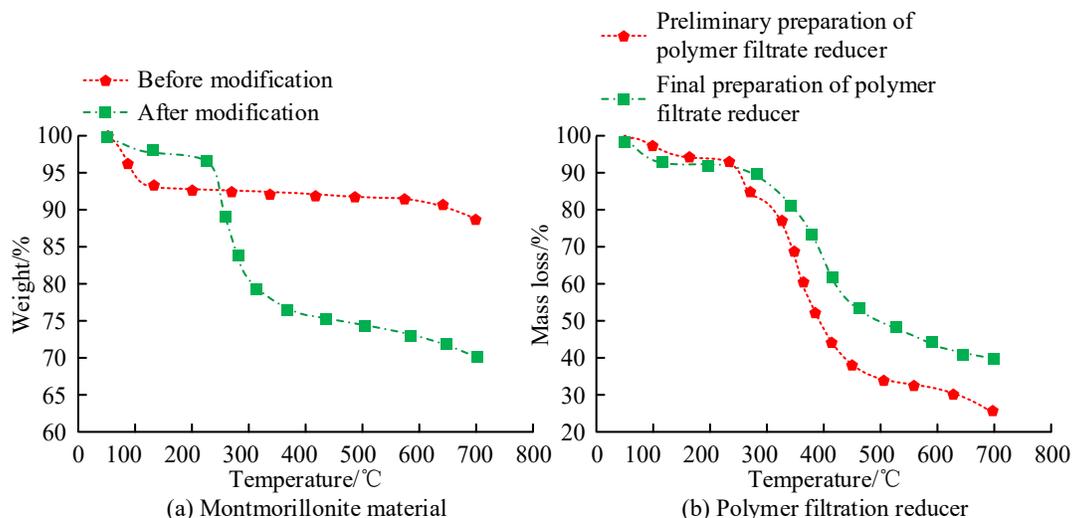


Figure 5. Thermogravimetric analysis of different materials.

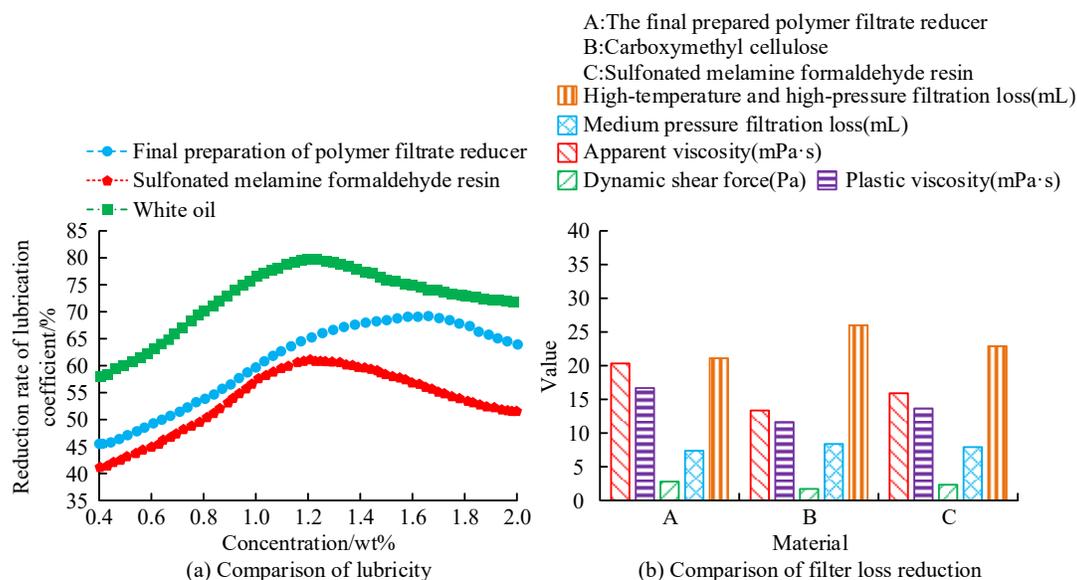


Figure 6. Comparison of lubricity and filtration properties of different filtrate reducers.

polymer filtrate reducers were 12.57% and 19.58%, respectively (Figure 5b). This result was because the addition of montmorillonite layers slowed down the internal heat conduction of the polymer and increased the difficulty of polymer decomposition.

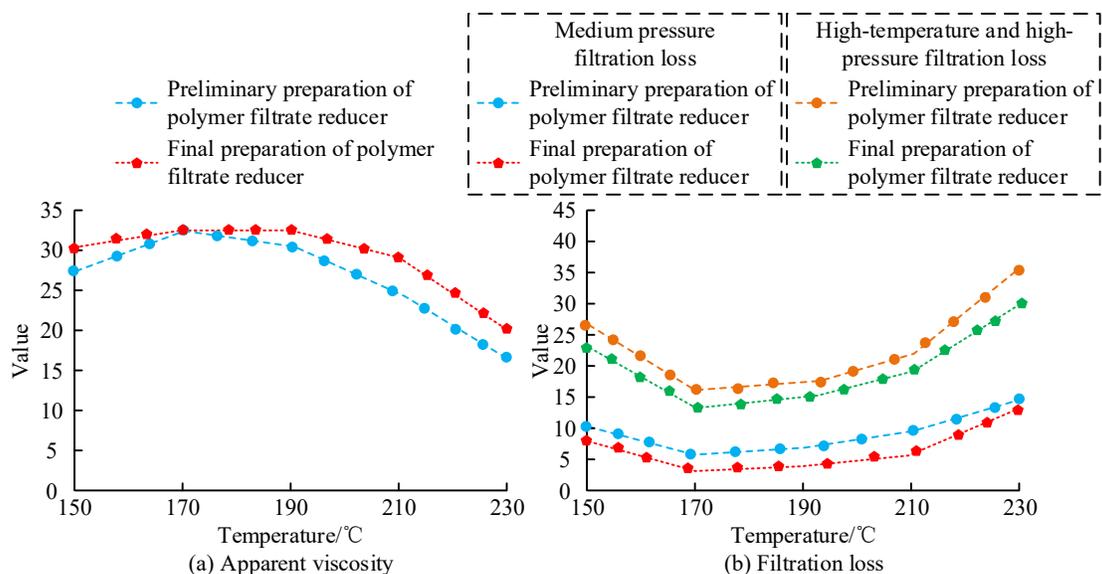
Performance evaluation of polymer materials

To further evaluate the performance of polymer filtrate reducers, sulfonated melamine

formaldehyde resin and white oil were selected to compare lubricity. Carboxymethyl cellulose and sulfonated melamine formaldehyde resin were used to compare filtrate. The results of lubricity comparison showed that, as the concentration of the filtrate reducer increased, the corresponding lubrication coefficients increased first and then decreased. The maximum reduction rates of lubrication coefficient for polymer filtrate reducer, white oil,

Table 1. The effect of the final dosage of polymer filtrate reducer on the performance of drilling fluid.

Index	Material dosage (%)					
	0	0.6	1.2	1.8	2.4	3.0
Apparent viscosity (mPa·s)	5.02	17.05	25.03	31.00	32.55	35.00
Plastic viscosity (mPa·s)	2.71	13.75	22.42	25.68	27.70	30.00
Dynamic shear force (Pa)	2.31	3.30	2.61	5.32	4.85	5.00
MPFL (mL)	20.00	15.00	9.13	5.00	5.00	6.77
HTHP filtration loss (mL)	60.00	35.00	22.34	17.23	15.08	19.15

**Figure 7.** The temperature resistance test results of the final preparation of polymer filtrate reducer.

and sulfonated melamine formaldehyde resin were 69.21%, 79.98%, and 61.69%, respectively (Figure 6a). The designed polymer filtrate reducers had a good lubricating effect. The apparent viscosity of the designed polymer filtrate reducer, carboxymethyl cellulose, and sulfonated melamine formaldehyde resin were 20.5 mPa·s, 13.75 mPa·s, and 15.87 mPa·s, respectively. The apparent viscosity of the designed polymer filtrate reducer exceeded that of the other materials, while it also had higher plastic viscosity and dynamic shear force than the other materials. In addition, the medium pressure filtration loss of the three materials was relatively close with HTHP filtration losses of 20.78 mL, 26.07 mL, and 22.65 mL, respectively (Figure 6b). The designed polymer filtrate reducer had great application value and good

fluid loss reduction and lubrication properties. The effect of the final dosage of polymer filtrate reducer on the drilling fluid demonstrated that, as the final amount of polymer filtrate reducer increased, the apparent viscosity, plastic viscosity, and dynamic shear force of the DFS all increased synchronously, while the corresponding medium pressure filtration loss and HTHP filtration loss first decreased and then increased. The material addition corresponding to the turning point of the medium pressure filtration loss and HTHP filtration loss was 2.4%, indicating that the filtration reduction performance of the DFS was better at this time. When the material dosage was 2.4%, the apparent viscosity, plastic viscosity, and dynamic shear force of the DFS were 32.55 mPa·s, 27.70 mPa·s, and 4.85 Pa, respectively (Table 1). The

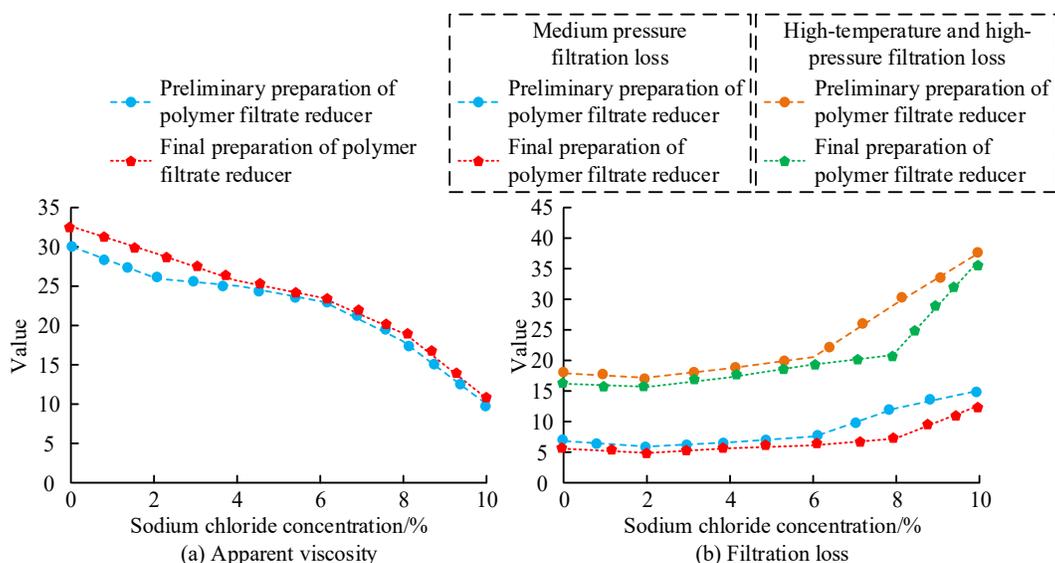


Figure 8. Salt resistance test results of the final preparation of polymer filtrate reducer.

results suggested that, when the material dosage reached a certain level, the DFS had better performance. The temperature resistance test results of the final prepared polymer filtrate reducer showed that, as the temperature increased, the apparent viscosity values of the preliminary and final polymer filtrate reducers first increased and then decreased, and the temperatures corresponding to the turning points were 170°C and 190°C, respectively. In addition, the apparent viscosity corresponding to both materials at the turning point temperature was 32.4 mPa·s (Figure 7a). The change of both materials in the medium pressure filtration loss and HTHP filtration loss first decreased and then increased with the increase of temperature. The two types of filtration losses of the initially prepared polymer filtration reducer were always higher than those of the final prepared material. At 210°C, the medium pressure filtration loss of the two materials was 9.27 mL and 6.98 mL, respectively, and the HTHP filtration losses were 21.86 mL and 19.54 mL, respectively (Figure 7b). The viscosity, filtration reducing ability, and temperature resistance of the final prepared polymer filtrate reducer were greater than those of the initially prepared material. The salt resistance test results of the final prepared polymer filtrate reducer showed that, as the

sodium chloride concentration increased, the apparent viscosity of the initially prepared polymer filtrate reducer remained consistently lower than that of the final prepared material. When the sodium chloride concentration was 10%, the apparent viscosity of the two materials was 10.0 mPa·s and 10.76 mPa·s, respectively (Figure 8a), which indicated that the viscosity of the final prepared material was stronger and could better reduce the filtration loss of the material. As the sodium chloride concentration increased, the medium pressure filtration loss and HTHP filtration loss of both materials first decreased and then increased, and the filtration loss of the final polymer filtrate reducer was always lower than that of the initial preparation material. The turning points of the two materials' filtration loss trends corresponded to a sodium chloride concentration of 2%. At this point, the initial medium pressure filtration loss and HTHP filtration loss of the prepared materials were 5.45 mL and 17.28 mL, respectively. The final medium pressure filtration loss and HTHP filtration loss of the prepared materials were 5.01 mL and 16.98 mL, respectively (Figure 8b). The final prepared polymer filtrate reducer had good salt resistance.

Evaluation of water-based drilling fluid system

Table 2. Evaluation results of water-based drilling fluid system.

Materials	ID.	Apparent viscosity (mPa·s)	Plastic viscosity (mPa·s)	Dynamic shear force (Pa)	MPFL (mL)	HTHP filtration loss (mL)
Sulfonated material ratio	S1	64	50	14	10.9	31.9
	S2	66	52	14	9.1	28.5
	S3	69	54	15	7.7	22.7
	S4	70	56	14	9.9	28.1
	S5	71	58	13	8.7	25.3
	S6	76	61	15	11.3	35.5
Lubricant	L1	67	55	12	10.7	31.5
	L2	68	55	13	9.9	27.1
	L3	69	56	13	8.5	25.9
	L4	72	58	14	7.3	19.9
	L5	70	57	13	7.7	22.7
	L6	72	59	13	7.9	23.8
Plugging agents	P1	72	56	16	8.1	21.1
	P2	73	56	17	7.3	19.9
	P3	73	56	17	7.1	18.5
	P4	72	56	16	7.3	19.3
	P5	72	57	15	7.9	20.5
Optimization		79	62	17	5.3	14.3

To optimize the water-based DFS scheme, the drilling fluid performance corresponding to each scheme was tested including apparent viscosity, plastic viscosity, dynamic shear force, medium pressure filtration loss, and HTHP filtration loss. The results showed that, in the sulfonated material ratio, the apparent viscosity, plastic viscosity, and dynamic shear force corresponding to different schemes were not significantly different (Table 2). Therefore, the selection of the optimal scheme mainly depended on the filtration loss. The medium pressure filtration loss and HTHP filtration loss corresponding to the S3 were 7.7 mL and 22.7 mL, respectively, which were lower than other schemes. Therefore, the total amount of sulfonated phenolic resin and sulfonated brown coal resin in the water-based DFS was 5.0%. In the selection of plugging agent materials, the L4 scheme had a smaller filtration loss. Meanwhile, in the selection of lubricant materials, the P3 scheme had a smaller filtration loss. Therefore, in the final water-based DFS, the dosages of the two plugging agents were 3.0% and 2.0%, respectively, and the lubricant dosage was 1.2%. The medium pressure filtration loss and HTHP filtration loss of the final solution were 5.3 mL and 14.3 mL, respectively, demonstrating excellent performance.

Conclusion

A filtration reducer incorporating modified montmorillonite nanomaterials was proposed in this research to construct the high-performance water-based DFS, while the other materials in the drilling fluid were also optimized. The results showed that, when the amount of modified montmorillonite nanomaterials was 1.1 wt%, it had stronger temperature resistance. When the designed filtrate reducer dosage was 2.4%, the viscosity, filtration capacity, and temperature resistance of the designed filtrate reducer were greater than those of the initially prepared material. The medium pressure filtration loss and HTHP filtration loss of the water-based DFS after material selection were 5.3 mL and 14.3 mL, respectively, indicating excellent performance. The designed filtrate reducer and water-based DFS had good performance and could reduce the amount of fluid loss. The lack of economic analysis of filtrate reducers was an important consideration for industrial applications. Future research should be improved from the perspectives of production costs, application costs, and economic benefits. Meanwhile, the study reflected the impact of multiple different single factors on materials without involving the

combined effects of factors. Future research should also improve this aspect.

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