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Development and Application of Eco-Friendly Micro-Nano Filtrate Reducers and High-Performance Water-Based Drilling Fluids

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Abstract: The utilization of oil-based drilling fluids is a significant technical approach for drilling in ultra-deep, unconventional, and other complex hydrocarbon reservoirs. However, these fluids present notable disadvantages, including high preparation costs and environmental pollution. There is an urgent need to develop an eco-friendly, high-performance water-based drilling fluid system suitable for complex geological conditions to support the exploration and development of oil and gas under deep, challenging, and unconventional conditions. Addressing the current issue where polymer filtrate reducers cannot simultaneously achieve temperature resistance, salt resistance, and environmental performance, a novel organic/inorganic composite micro-nano filtrate reducer (MNFR) was developed using inverse emulsion polymerization. The MNFR has a D_{50} particle size of $1.313\mu m$, withstands temperatures up to $200^{\circ}C$, resists saturated NaCl brine, and exhibits an EC_{50} biotoxicity value of $86700 mg/L$. Furthermore, a high-temperature-resistant (up to $200^{\circ}C$) eco-friendly high-performance drilling fluid system (HBHP) was constructed, demonstrating excellent rheological and filtration properties, with a high temperature and high pressure (HTHP) filtration volume of only 7.6 mL and an EC_{50} biotoxicity value of $54300 mg/L$. It also shows outstanding plugging, anti-collapse, and hydration inhibition properties. The HBHP system has been applied in three wells in the Shengli oilfield, with no complex situations related to wellbore stability occurring during field operations, thus providing technical support for the green development of complex hydrocarbon reservoirs such as deep, ultra-deep, offshore deepwater, and unconventional formations.

Keywords: Water-based drilling fluid; High performance; Environmental friendly; Filtrate reducer; Micro-nano scale; Temperature and salt resistance

1 Introduction

Drilling fluids, often referred to as the "blood" of drilling engineering, play a crucial role in ensuring safe, efficient, high-quality, and environmentally friendly drilling operations. However, waste drilling fluids are a significant pollutant in drilling engineering [1–3], characterized by large volumes, substantial environmental hazards, and considerable challenges in achieving harmless disposal. With the implementation of stringent environmental laws and regulations, the environmental protection situation in petroleum engineering is becoming increasingly severe. It is required that waste drilling fluids, oil-containing cuttings and other drilling fluid waste that do not meet discharge standards during oil and gas development be collected and centrally treated [4–6] to achieve resource utilization or harmless disposal. Failure to do so results in substantial environmental protection taxes being imposed.

Major international oil service companies such as Schlumberger, Baker Hughes, and Halliburton have developed high-performance, eco-friendly water-based drilling fluid systems [7–10]. The core additives in the latest generation of these systems are micro-nano composite materials, which provide functions such as temperature and salt resistance, filtration loss reduction, and viscosity enhancement. These formulations simplify drilling fluid compositions to five or six core additives, making them easy to adjust and capable of meeting the discharge standards of the Gulf of Mexico [11]. In China, high-performance water-based drilling fluid systems such as polyamine, aluminum amine, "water-for-oil" for shale gas, alkyl glycoside, and cationic silico-fluoropolyester have been developed [12–14]. However, due to a lack of efficient, eco-friendly additives, the challenges of performance regulation and environmental friendliness have not been fully addressed, necessitating targeted research on eco-friendly, high-performance water-based drilling fluid systems and their supporting additives.

Organic/inorganic nanocomposites combine the rigidity, size stability, and thermal stability of inorganic nanoparticles with the toughness, processability, and node performance of polymers [15–18]. These materials leverage the synergistic and complementary effects between their components to achieve superior properties, meeting the multifaceted requirements for drilling fluid additives such as temperature and salt resistance, filtration loss reduction, and flow pattern regulation [19, 20]. Consequently, they hold significant potential for application in the field of drilling fluid additives [21].

In response to the complex formulations, excessive biotoxicity, and heavy metal content of current water-based drilling fluid systems, MNFR was designed and developed. Subsequently, an eco-friendly, high-performance water-based drilling fluid system (HBHP) was constructed and successfully applied in the field. This system significantly improved the performance indicators of drilling fluids, achieved non-toxic and green drilling operations, and provided technical support for the green development of complex hydrocarbon reservoirs, including deep, ultra-deep, offshore deepwater, and unconventional formations.

2 Materials and Methods

2.1 Development of MNFR

2.1.1 Molecular structure design

The surface of nano-calcium carbonate was modified using the silane coupling agent KH570. Through inverse emulsion polymerization, sodium vinyl sulfonate (SVS), N-vinylpyrrolidone (NVP), acrylamide (AM), and 2-acrylamido-2-methylpropane sulfonic acid (AMPS) monomers were grafted onto the surface of nano-calcium carbonate, resulting in the novel organic-inorganic composite MNFR. The introduction of hydrophobic groups with hydrophobic association characteristics facilitated the mutual association and self-assembly of hydrophobic groups within the molecular chains of MNFR. This maintained a relatively intact micro-crosslinked spatial network structure under harsh conditions such as high temperature and saturated brine, thereby enhancing the colloidal stability of the drilling fluid. The presence of numerous polar adsorption groups in the molecular chains prevented the aggregation of clay particles and improved the particle size distribution of clay particles. This contributed to the formation of a dense mud cake with a membrane-like structure, meeting the multifaceted requirements of temperature resistance, salt tolerance, and filtration loss reduction.

2.1.2 Preparation of MNFR

(a) Experimental reagents. SVS, NVP, AM, AMPS, silane coupling agent KH570, ammonium persulfate, sodium bisulfite, acetone, and anhydrous ethanol are all of analytical grade. Emulsifiers A and B were prepared in the laboratory. White oil and nano-calcium carbonate are of industrial grade.

(b) Preparation method. A certain amount of emulsifier A and emulsifier B was added to white oil, stirred, and dissolved to obtain a uniform transparent liquid. A suitable amount of deionized water was taken, and SVS, NVP, AM, and AMPS were slowly added in sequence and stirred until a uniform transparent solution was obtained. Under continuous stirring, KH570-modified nano-calcium carbonate was slowly added and then slowly dripped into the continuous phase of white oil. Nitrogen gas was introduced, and the temperature was increased to 60-70°C. The initiator was added, and the reaction was allowed to proceed for 3-4 hours. After the reaction was completed, the product was repeatedly washed with anhydrous ethanol and dried to obtain the final product, namely the MNFR.

2.1.3 Structural characterization

(a) Infrared spectroscopy analysis. As shown in Figure 1, the infrared spectrum of nano-calcium carbonate exhibits a $C = O$ stretching vibration peak at $1790.04\ cm^{-1}$ and an asymmetric $C - O$ stretching vibration peak at $1426.93\ cm^{-1}$. In the infrared spectrum of MNFR , the $N - H$ stretching vibration peak appears at $3382\ cm^{-1}$, the -OH stretching vibration peak at $3199.38\ cm^{-1}$, the methylene stretching vibration peak at $2947.75\ cm^{-1}$, the $C = O$ absorption peak in the amide group at $1664.29\ cm^{-1}$, and the characteristic absorption peak of $-SO_3{}^{2-}$ at $1128.17\ cm^{-1}$. These findings indicate that the functional groups of the reactive monomers were successfully grafted onto the surface of nano-calcium carbonate, meeting the requirements of the molecular structure design.

(b) Morphological characterization. A 0.5% MNFR suspension was prepared using deionized water and ultrasonically dispersed for 10 minutes. The laser particle size analysis results indicate that the particle size of the MNFR is within the micro-nano scale, displaying a multimodal distribution with a D_{50} particle size of $1.313\mu m$ and a D_{90} particle size of 7.279 um . As shown in Figure 2, the MNFR comprises numerous square or near-spherical particles with diameters ranging from 200 to 300 nm , as well as some micron-sized particle aggregates. The irregular nano-calcium carbonate particles were coated with a polymer layer, forming MNFR with a core-shell structure.

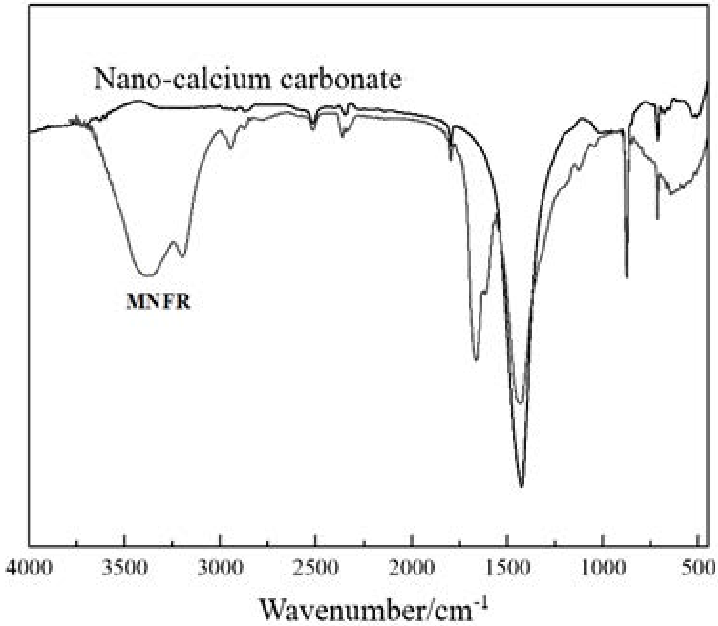

Figure 1. Diagram of Fourier-transform infrared spectroscopy (FT-IR)

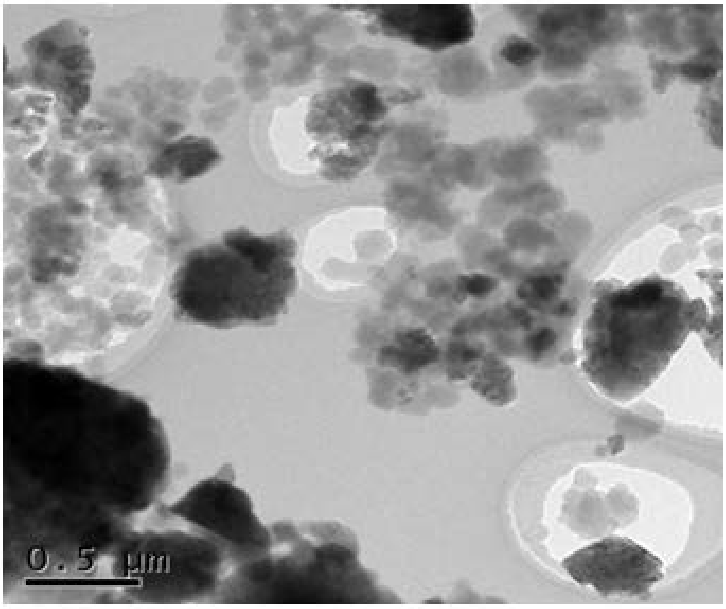

Figure 2. Transmission electron microscope (TEM) images

2.2 Construction of the Eco-Friendly High-Performance Water-Based Drilling Fluid System (HBHP)

2.2.1 Optimization of drilling fluid formulation

Based on the MNFR, the eco-friendly viscosifier HBVF was selected as a rheology modifier. The eco-friendly shale inhibitor HBHIB and the eco-friendly plugging and anti-collapse agent HBWA were used to enhance hydration inhibition and plugging performance, thereby improving wellbore stability. The high-temperature-resistant eco-friendly lubricant HBLU was chosen to enhance the extreme pressure lubrication performance of the drilling fluid. According to the requirements of on-site drilling fluid performance, the eco-friendly deflocculant HBVR was used to regulate the viscosity and shear force of the drilling fluid system, ensuring appropriate rheological properties of the drilling fluid. Evaluation results indicate that the EC_{50} value for the biological toxicity of the selected key additives is $\geq 30000\mathrm{mg/L}$, and the biodegradability $(\mathrm{BOD}_5/\mathrm{COD}_{\mathrm{Cr}})$ is $\geq 10\%$, meeting the environmental performance requirements of being non-toxic and biodegradable.

Compatibility evaluation experiments were conducted to optimize the optimal dosage of core additives, including

MNFR, HBVF, HBHIB, HBWA, and HBLU. This led to the construction of a high-temperature-resistant (up to $200^{\circ}C$) eco-friendly high-performance water-based drilling fluid system (HBHP). The optimized formulation is as follows: 4% bentonite slurry $+0.2\%$ eco-friendly viscosifier HBVF $+1.8\%$ MNFR $+3.0\%$ eco-friendly anti-collapse agent HBWA $+2.0\%$ eco-friendly inhibitor HBHIB $+2.0\%$ nano plugging agent SDSP $+2.5\%$ high-temperature-resistant eco-friendly lubricant $HBLU + 6\%NaCl + 5\%KCl$ (weighted with barite to $1.5\ g/cm^3$).

2.2.2 Performance evaluation of drilling fluids

The performance of the eco-friendly high-performance water-based drilling fluid system (HBHP) was tested following the API RP 13B-1-2009 *Recommended Practice for Field Testing Water-Based Drilling Fluids*and GB/T 16783.1-2014 *Petroleum and Natural Gas Industries—Field Testing of Drilling Fluids—Part 1: Water-Based Fluids*. The tests assessed the rheological properties, filtration, contamination resistance, hydration inhibition, and plugging and anti-collapse performance of the HBHP system.

The environmental performance of the HBHP system was evaluated according to SY/T 6788-2010 *Technical Evaluation Methods for Environmental Protection of Water-Soluble Oilfield Chemicals*and GB 18420.1-2009 *Toxicity to Marine Organisms from Offshore Exploration and Production Effluents—Part 1: Classification/Part 2: Test Methods*.

3 Results and Discussion

3.1 Performance Evaluation of MNFR

3.1.1 Performance of the filtrate reducer

The base slurry, which is 4% pre-hydrated bentonite slurry, was used to test the rheological and filtration properties before and after hot rolling at 160°C for 16 hours. As shown in Table 1, the addition of the MNFR significantly reduced the API filtration volume of the test slurry. Furthermore, as the dosage of MNFR increased, the API filtration volume decreased correspondingly. When the dosage reached 1%, the API filtration volume after hot rolling at 160°C for 16 hours was only 9.2 mL (less than 10 mL), indicating excellent filtration reduction performance.

Table 1. Evaluation results of MNFR's filtration reduction performance

Formula	Condition	$AV(mPa\cdot s)$	$PV(mPa\cdot s)$	$YP(Pa)$	$FL_{API}(mL)$	pH
0	Before hot rolling	7.0	5.0	2.0	25.4	10
	After hot rolling	3.5	2.5	1.0	36.6	10
0.5	before hot rolling	18.0	11.0	7.0	9.0	10
	After hot rolling	20.0	18.0	2.0	16.8	10
1.0	Before hot rolling	32.5	17.0	15.5	8.0	10
	After hot rolling	28.5	24.0	4.5	9.2	10
2.0	Before hot rolling	62.5	31.0	31.5	6.8	10
	After hot rolling	67.5	37.5	30.0	6.4	10
3.0	Before hot rolling	98.5	54.0	44.5	6.0	10
	After hot rolling	105.0	60.0	45.0	6.0	10

3.1.2 Temperature resistance

The base slurry, which is 4% pre-hydrated bentonite slurry, with the addition of 1.0% MNFR, was used to test the rheological and filtration properties before and after hot rolling at different temperatures. As shown in Table 2, the API filtration volume slightly increased with the rise in hot rolling temperature. After hot rolling at 200°C, the API filtration volume was only 12.0 mL, indicating that the MNFR can withstand temperatures up to 200°C, meeting the filtration reduction requirements for high-temperature deep well drilling fluids.

Table 2. Evaluation results of MNFR's temperature resistance

Aging Condition	$AV(mPa\cdot s)$	$PV(mPa\cdot s)$	$YP(Pa)$	$FL_{API}\ (mL)$	pH
Before hot rolling	32.5	17.0	15.5	8.0	10
After hot rolling at $160^{\circ}C$	28.5	24.0	4.5	9.2	10
After hot rolling at $180^{\circ}C$	25.0	19.0	6.0	9.4	10
After hot rolling at $200^{\circ}C$	21.0	17.0	4.0	11.2	10

3.1.3 Salt resistance

The base slurry, which is 4% pre-hydrated bentonite slurry, with the addition of 1.0% MNFR, was used to test the rheological and filtration properties before and after hot rolling at 200°C for 16 hours under different concentrations of NaCl. As shown in Table 3, the API filtration volume significantly decreased before and after hot rolling at 200°C for 16 hours as the NaCl concentration increased. When the NaCl concentration reached saturation, the API filtration volume was only 42.8 mL, indicating that the MNFR possesses excellent salt resistance and can withstand saturated NaCl contamination at high temperatures of 200°C.

3.1.4 Environmental performance

The environmental performance of MNFR was evaluated according to the petroleum and natural gas industry standard SY/T 6788-2010, using indicators such as biological toxicity (EC_{50}), biodegradability (BOD_5/COD_{cx}), and heavy metal content. The analysis indicates that the MNFR has a biological toxicity EC_{50} value of $86700 mg/L$ and a biodegradability BOD_5/COD_{ct} value of 17.21%, meeting the environmental requirements of being non-toxic and biodegradable.

3.2 Performance Evaluation of the Drilling Fluid System HBHP

3.2.1 Rheological and filtration properties

Table 3 presents the evaluation results of the rheological and filtration properties of the HBHP system. The analysis indicates that the HBHP system maintained relatively stable rheological and filtration properties before and after hot rolling at 200°C for 16 hours. The plastic viscosity remained within 35 mPa.s, the yield point within 20 Pa, and the filtration control performance was satisfactory. After hot rolling at 200°C for 16 hours, the API filtration volume was 2.4 mL, and the HTHP filtration volume was 7.6 mL. These results demonstrate that the HBHP system can withstand temperatures up to 200°C, meeting the requirements for deep well drilling operations in the field.

Table 3. Evaluation results of the rheological and filtration properties of the HBHP system

Experimental Condition	$AV(mPa, s)$	$PV(mPas)$	$YP(Pa)$	$Gel(Pa)$	$FL_{spI}(mL)$	FL_{HTHP} (mL)	pH
Before hot rolling	52.5	34.5	18.0	6.0/10.5	2.8	-	10
After hot rolling at 200°C	45.5	32.0	13.5	4.5/8.0	2.4	7.6	9.5

3.2.2 Contamination resistance

The HBHP system maintained good rheological and filtration properties before and after hot rolling with the addition of $20\% NaCl$ and $1\% CaCl_2$. The changes in viscosity and yield point were minimal, and the API filtration volume remained less than 5 mL , demonstrating excellent resistance to salt and calcium contamination. After the addition of 10% evaluation soil, the viscosity and yield point of the HBHP system increased slightly but still maintained satisfactory rheological and filtration properties, indicating good resistance to poor soil contamination.

3.2.3 Environmental performance

The environmental performance of the HBHP system was evaluated according to the petroleum and natural gas industry standard SY/T 6788-2010. The test results show that the HBHP system exhibits excellent environmental performance, with a biological toxicity EC_{50} value of $54300 mg/L$, low heavy metal content, and a light grey color. Additionally, according to the national standard *Toxicity to Marine Organisms from Offshore Exploration and Production Effluents-Part 1: Classification/Part 2: Test Methods*(GB 18420.1-2009), the 96-hour biological toxicity LC_{50} value is greater than 30000 mg/L, meeting the biological toxicity requirements for China's first and second-class sea areas.

3.2.4 Hydration inhibition performance

The hydration inhibition performance of the HBHP system was evaluated using shale expansion and rolling dispersion tests, with linear expansion rate and rolling recovery rate as evaluation indicators. Simulated rock samples prepared from sodium bentonite were tested for linear expansion rate, with fresh water as a control. The analysis indicates that the simulated rock samples showed a rapid expansion rate in fresh water, with an 8-hour linear expansion rate exceeding 27%. In contrast, the linear expansion rate in the HBHP system was significantly reduced, decreasing to 3.75% after 8 hours.

Rolling dispersion tests were conducted using mud shale samples (6-10 mesh) at 77°C for 16 hours, with fresh water as a control, to evaluate the rolling recovery rate of the HBHP system. The analysis showed that the rolling recovery rate of the mud shale samples in fresh water was only 24.91%, whereas it significantly increased to 95.67% in the HBHP system. These results indicate that the HBHP system possesses excellent capabilities for inhibiting the hydration, dispersion, and expansion of clays.

3.2.5 Plugging and anti-collapse performance

The plugging and anti-collapse performance of the HBHP system were evaluated using a permeability plugging apparatus (PPA) with ultra-low permeability (400 mD) sand discs. The comparison results are shown in Figure 3). The PPA filtration volume of the field drilling fluid system is relatively high at 42.2 mL , whereas the PPA filtration volume of the HBHP system significantly reduces to 13.0 mL . Additionally, the instantaneous filtration volume of the field polysulfonate drilling fluid system is 6.26 mL , with a static filtration rate of $0.57\ \mathrm{mL/min^{1/2}}$. In comparison, the HBHP system exhibits an instantaneous filtration volume of 1.77 mL and a static filtration rate of $0.18\ \mathrm{mL/min^{1/2}}$, demonstrating superior plugging and anti-collapse performance.

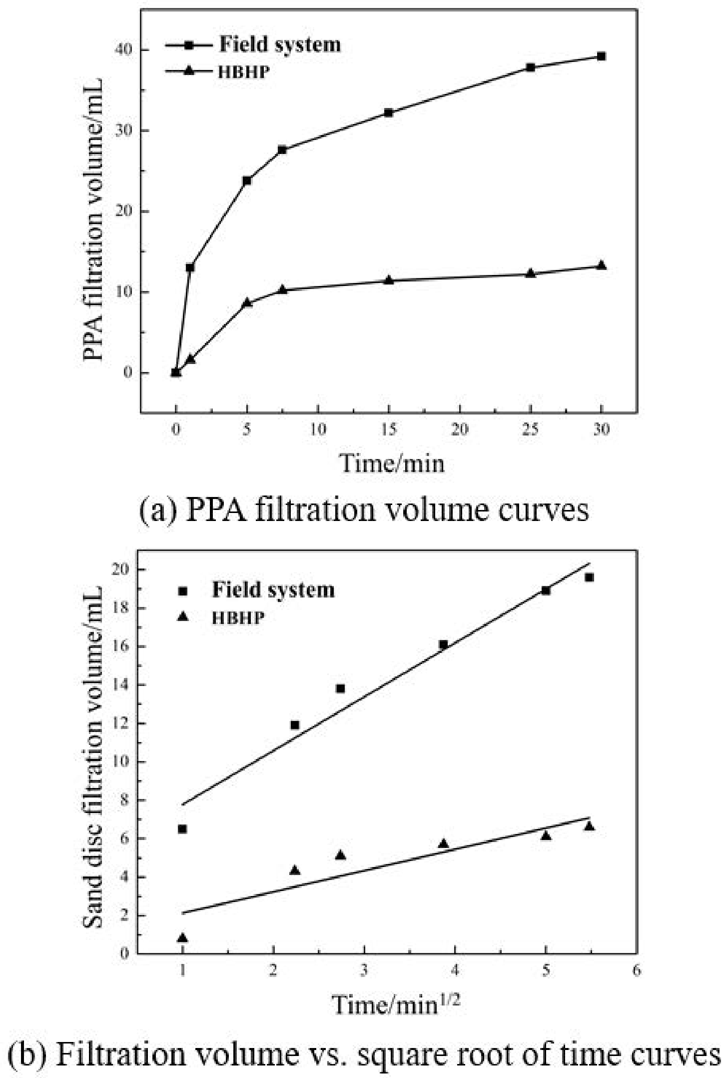

Figure 3. Results of permeability plugging tests

3.3 Field Application and Performance Evaluation

The eco-friendly high-performance water-based drilling fluid system (HBHP) has been applied in three wells in the Shengli oilfield. During field operations, no complex situations related to wellbore stability occurred, demonstrating excellent field application results. The performance indicators of the drilling fluid were significantly improved, achieving a green and non-toxic drilling process.

The BS-X well, a key exploratory well deployed in the northern part of the Jiyang Depression, was designed to reach a depth of 6518 meters as a directional well with a maximum horizontal displacement of 3080 meters. The anticipated bottom hole temperature is approximately 190°C, and the well has a four-section wellbore structure. The main technical challenges for the drilling fluid in the BS-X well include high formation temperatures in the fourth section, making the regulation of thermal stability, rheology, and filtration properties difficult. The Shahejie Formation exhibits hard, brittle shale, oil shale, and calcareous mudstone with well-developed bedding and microfractures, making it prone to collapse and block falling. Additionally, stringent environmental pollution control requirements necessitate the use of an eco-friendly water-based drilling fluid system for field drilling operations.

Given these technical challenges and environmental requirements, the HBHP system was employed for the fourth section of the BS-X well. Field application results indicate that the HBHP system maintains stable rheology and filtration properties, exhibits excellent extreme pressure lubrication and plugging and anti-collapse performance, and possesses strong contamination resistance. The system is easy to handle and maintain on site, with no complex situations related to wellbore stability occurring. Tripping operations are smooth, and the logging success rate is 100%. Post-drilling environmental testing shows that the biological toxicity EC50 value of the drilling fluid exceeds 30000 mg/L, indicating excellent environmental performance and meeting the environmental requirements for drilling fluids.

4 Conclusions

A novel organic/inorganic composite MNFR was developed, with a D_{50} particle size of $1.313\mu\mathrm{m}$, capable of withstanding temperatures up to $200^\circ\mathrm{C}$ and resisting saturated NaCl brine. The biological toxicity EC_{50} value was determined to be $86700\mathrm{mg/L}$. Additionally, an eco-friendly high-performance water-based drilling fluid system (HBHP) was constructed, demonstrating a high-temperature resistance of up to $200^\circ\mathrm{C}$, with an HTHP filtration volume of only 7.6 mL and a biological toxicity EC_{50} value of $54300\mathrm{mg/L}$. During field operations, the HBHP system exhibited excellent high-temperature performance and stable rheological and filtration properties. No complex situations related to wellbore stability were encountered, significantly enhancing the environmental performance of the drilling fluid while ensuring its engineering performance. This system provides robust technical support for the green development of complex hydrocarbon reservoirs, including deep, ultra-deep, offshore deepwater, and unconventional formations.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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