



pH-Responsive Drilling Fluid With High-Temperature and High-Density Performance

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Conventional drilling fluids experience the problems of a low cleanup efficiency and oily drilled cutting disposal. To resolve these problems, a type of pH-responsive drilling fluid with a temperature resistance of up to 150°C resistance a density of 1.5 g/cm³ was prepared using mixed emulsifiers. Stable reversion from a water-in-oil (W/O) emulsion to an oil-in-water (O/W) and vice versa was realized. The results of light backscattering and sag stability suggested that the W/O and O/W emulsion-based drilling fluids showed excellent resistance to coalescence or sedimentation. There was no demulsification or barite sedimentation during emulsion reversion. W/O and O/W emulsions exhibited small droplet sizes and uniform distributions. The properties of the W/O and O/W emulsionbased drilling fluids were similar even after hot rolling at 150°C for 16 h. The reversible drilling fluid showed excellent resistance to contamination by saline water and drilled cuttings. The residual W/O emulsion-based drilling fluid was cleaned with acids at a high efficiency. The oil content of the drill cuttings generated by the W/O emulsion-based drilling fluid was reduced using simple acids. The improved reversion stability with high temperature and density resistance makes it possible for applicable reversible drilling fluid in deep or ultra-deep wells required by different geological formation.

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INTRODUCTION

Conventional oil-based drilling fluids typically have high emulsion stability and strong oil-wetting properties, which render contact drilling pipes, formation, and drill cuttings oil-wet (Nooripoor and Hashemi, 2020). In drilling procedures, strong oil-wetting properties are beneficial for improving shale formation stability and reducing downhole accidents. However, in follow-up cementing and completion procedures, strong oil-wetting properties make it difficult to efficiently remove the residual drilling fluid and filter cakes in a wellbore, which might result in poor cementing quality and production safety. In addition, it is expensive to dispose the oily drill cuttings that are produced by oil-based drilling fluids when they are circulated back to surfaces. Hence, maintaining the advantages of conventional drilling fluids while reducing their adverse effects is an emerging research topic.

Previous studies have proposed several methods to solve the follow-up problems associated with conventional oil-based drilling fluids, e.g., oil-based drilling fluid filter-cake removal (Bageri et al., 2019; Bageri et al., 2020), hole cleaning (Boyou et al., 2019; Guo et al., 2020), and waste oily drilled cutting processing (Sharma et al., 2020). However, these methods are expensive and complex, and



they may cause secondary pollution. Controlling the wettability of oil-based drilling fluids is a promising new solution.

Patel (1999) proposed the first reversible emulsion drilling fluid in 1999. Reversible drilling fluids and conventional oil-based drilling fluids are different in terms of pH-responsive emulsifiers (Patel, 1999; Patel et al., 1999; Patel and Ali, 2003). In drilling procedures, reversible drilling fluids are in the water-in-oil (W/O) emulsion phase, which provides advantages similar to those of conventional drilling fluids. In the follow-up cementing and completion stage, reversible drilling fluids are transformed into the oil-in-water (O/W) emulsion phase, which makes it easy to remove residual drilling fluids and filter cakes.

Reversible emulsions can be stimulated by magnetism (Brugger and Richtering, 2007; Chen et al., 2014), electricity (Quesada et al., 2013), temperature (Binks et al., 2005; Destribats et al., 2012), electrolytes (Cui et al., 2008; Cui et al., 2012; Ren et al., 2017; Xu et al., 2018), pH (Read et al., 2004; Fujii et al., 2005; Binks and Rodrigues, 2007; Haase et al., 2010; Dyab, 2012; Ghosn et al., 2017), and the CO₂/N₂ (Zhang et al., 2016; Shi et al., 2018; Guo and Zhang, 2019) or volume ratio (Popov et al., 2013). Among these, pHresponsive emulsions are one of the simplest and most readily implementable systems (Tang et al., 2015). Amine surfactants form responsive emulsions by transitioning from a surface-active substance (cationic ammonium) to a non-surface-active substance (neutral tertiary amine) upon exposure to CO₂/N₂ (Zhang et al., 2016), (Guo and Zhang, 2019), (Tang et al., 2015), (Shi et al., 2019). This transition depends on the pH change in the aqueous solution (pH = 5-10). However, current reversible drilling fluids stabilized by amine emulsifiers in oil fields have a typical temperature resistance of 120°C, and their density is lower than 1.2 g/cm (Dick et al., 2003; Ali et al., 2004; Bageri et al., 2019). These properties cannot meet the requirements of deep downhole environments. Furthermore, W/O drilling fluids have been extensively investigated, whereas O/W drilling fluids are difficult to use owing to their instability.

Even though numerous responsive emulsifiers have been developed, it is difficult to control the emulsion reversion stability

in high-temperature and high-pressure conditions. The breakdown of drilling fluids in downhole environments increases the risk of drilling accidents and decreases drilling safety. Therefore, the emulsion reversion stability and effectiveness can be used as a basis to develop a reversible drilling fluid that is, suitable for downhole environments. High-temperature resistance and high-density capacity require the reformulation of the reversible drilling fluid system.

In this study, we formulated a reversible drilling fluid with high temperature resistance and high density for deep or ultra-deep wells. Stable reversion from a water-in-oil (W/O) emulsion to an oil-in-water (O/W) and vice versa was realized. This work can provide guidance for the potential applications of reversible drilling fluids in wellbores.

MATERIALS AND METHODS

Experiments were performed using diesel oil purchased from a Shell gas station in Chengdu, Sichuan, China. The water used in the experiments was deionized using a Milli-Q reagent water system. A long-chain polyamine primary surfactant (hereinafter referred to as RPEM) was prepared according to a previously reported procedure (Zhang et al., 2013; Zhang et al., 2015). Sodium hydroxide (AR) and hydrochloric acid (35%, AR), which were obtained from Chengdu Kelong Reagent Plant, Chengdu, Sichuan, China, were used to adjust the pH of the solution. Organoclay, a filtration loss reducer, and a weight material (barite) were obtained from Chengdu Xiyouhuawei Science & Technology Co., Ltd. Surfactant flushing fluid was obtained from an oil field.

Preparation and Characterization of Emulsions

Emulsion Preparation

Equal volumes (150 ml) of the diesel oil phase containing RPEM and the aqueous phase (150 ml) were emulsified using a high-speed stirrer at 10,000 rpm for 30 min–1 h at room temperature

 $(25 \pm 5^{\circ}C)$. It should be noted that the surfactant concentrations mentioned in this paper refer to the content in the initial oil phase and are not based on the total volume. Emulsion drop size distributions were determined using a particle size distribution analyzer (LA-950V2, Horiba Scientific, China). The optical images of the emulsions were obtained *via* an inverted fluorescence digital microscope (AMG EVOSFL, Advanced Microscopy Group, USA). pH values for emulsions were detected by using PHS-3C.

Emulsion Reversion

The pH was adjusted by adding 1 M NaOH and/or 1 M HCl in a dropwise manner. As the addition of pH solution is minimal, its impact on the water-oil ratio is negligible. Immediately after emulsification, the emulsion type was determined by adding an emulsion drop to either pure oil or pure water. The emulsion was the O/W type if the drop dispersed in pure water and remained a drop in pure oil, and *vice versa* for the W/O type.

Emulsion Stability

The emulsions were transferred into stoppered and graduated glass vessels with inner diameters of 1.8 cm and lengths of 7 cm while maintaining the temperature at 25°C. For W/O emulsions, the downward movement of the oil–emulsion boundary was used as a measure of sedimentation and the change in the water–emulsion interface position was used as an indicator of coalescence. The extent of creaming in O/W emulsions was obtained by measuring the height of water resolved from the emulsions. The extent of sedimentation in O/W emulsions was obtained by measuring the height of oil resolved from the emulsions.

Conductivity

The conductivity of the emulsions was measured using a digital conductivity meter immediately after preparation. The emulsions with a high conductivity were classified as O/W emulsions or W/O/W emulsions and those with a low (immeasurable) conductivity (< 1 μ S cm⁻¹) were classified as W/O emulsions or O/W/O emulsions.

Demulsification Voltage (ES)

The ES values of the emulsions were determined using an electrical stability tester (DWY-2, Qingdao Tongchun Oil Instrument Co., Ltd., China) with an electrode distance of 1.55 ± 0.04 mm. The ES value is mainly used to measure the relative stability of W/O emulsions. A high electrical stability indicates high emulsion stability. The voltage required to break O/W emulsions is typically quite low (< 10 V).

Characterization of Reversible Drilling Fluid

The emulsions were thermally aged in a rotary oven at different temperatures for 16 h to model actual drilling operation conditions, in accordance with the American Petroleum Institute (API) SPEC 13A and RP 13B-2 standards for drilling fluids.

HTHP Filtration Loss

The HTHP filtration volume (FL_{HTHP}) was collected using a high-temperature and high-pressure filtration apparatus (GGS42,

Qingdao Tongchun Machinery Petroleum instrument, China) by applying a pressure of 4.2 ± 0.03 MPa at a given temperature.

Rheology Test

The rheological parameters, i.e., apparent viscosity (AV), plastic viscosity (PV), and yield point (YP), were obtained using a six-speed rotational viscometer (ZNN-D6, Qingdao Tongchun Petroleum Instrument Co., China) at 600 and 300 rpm. They were calculated as follows:

$$AV = \frac{1}{2}\theta_{600} (\text{mPa} \cdot \text{s})$$
$$PV = \theta_{600} - \theta_{300} (\text{mPa} \cdot \text{s})$$
$$YP = \frac{1}{2} (\theta_{600} - PV) (\text{Pa})$$

 θ_{600} and θ_{300} denote the dial readings on the viscometer at 600 and 300 rpm, respectively. To reach steady conditions, the fluids were stirred for 10 min prior to rheological measurements. G_{10s}/G_{10min} were the gel strength obtained at 10 s and 10 min for dial readings at 3 rpm. The experimental error for viscosity was always below ±5%.

Filter-Cake Cleanup

Filter cakes were built by immersing the rotational viscometer in the drilling fluids for 1 h. Then, the viscometer cylinders were soaked in washing fluids for 10 min to remove the filter cakes on their surfaces. The cleanup efficiency of the filter cakes was calculated by recording the differences in the weights of the cylinders before and after immersion.

Drilled Cuttings

Oily drill cuttings at 10–40 mesh, obtained from shale formation, were prepared by hot rolling the drilling fluids at 150°C for 16 h and sieving at room temperature. The oil content of the drill cuttings was measured using a Soxhlet extractor and refluxed for 12 h using petroleum ether.

Sag Test

The densities of the top and bottom sections of the drilling fluids were measured after static standing for 24 h. The sag factor (SF) was calculated as follows: SF = $\frac{\rho_{battom}}{\rho_{ros} + \rho_{battom}}$.

Emulsion Stability

Emulsion destabilization results from coalescence or flocculation was measured on a Multiple light scattering analyzer TURBISCAN MA 2000 at room temperature ($25 \pm 5^{\circ}$ C).

RESULTS AND DISCUSSION

Emulsification Efficiency

The pH-dependent behavior of the emulsion stabilized by only RPEM (2 wt%) was investigated in detail. **Figure 1** shows the variation in the ES value, conductivity, pH, and continuous phase resolution of the pH-responsive emulsion on the addition of the acid.

A stable W/O emulsion is generated by RPEM. As the acid concentration increases, the ES value significantly decreases from





emulsion during reversion. (C) Continuous phase resolution of emulsion. (D) Images obtained 2 h after the emulsions were prepared.







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Hot-rolling temperature/°C	Emulsion type	ES	AV	PV	YP/Pa	G _{10s} /G _{10min}	FL _{HTHP}
		/V	/mPa⋅s	/mPa⋅s			/mL
100	W/O	467	56.5	45	11.5	3.0/3.5	3.8
	O/W	4	58.5	47	11.5	3.5/4.0	4.2
120	W/O	444	52.0	40	12	2.5/3.5	3.6
	O/W	3	53.5	41	12.5	3.0/3.5	6.2
150	W/O	422	51.5	40	11.5	3.0/4.0	4.8
	O/W	6	50.5	39	11.5	3.5/4.5	8.2
180	W/O	354	30.5	22	8.5	1.0/1.5	16.8
	O/W	6	38.5	30	8.5	1.5/1.5	18.4

TABLE 2 | Variation in density of reversible drilling fluid.

Density	W/O emulsion-based drilling fluid	O/W emulsion-based drilling fluid
Top section (g/cm ³)	1.48	1.47
Bottom section (g/cm ³)	1.53	1.51
Density variation (g/cm ³)	0.05	0.04
SF	0.508	0.507

TABLE 3 | Properties of reversible drilling fluid contaminated by saline water.

Salinity	Emulsion type	ES/V	AV/mPa⋅s	PV/mPa⋅s	YP/Pa	G _{10s} /G _{10min}	FL _{HTHP} /mL
0	W/O	422	51.5	40	11.5	3.0/4.0	4.8
	O/W	6	50.5	39	11.5	3.5/4.5	8.2
5%CaSO ₄	W/O	419	53.5	41	12.5	3.0/3.5	5.2
	O/W	6	49.5	39	10.5	3.0/4.0	6.4
5%NaCl	W/O	427	59.0	47	12.0	2.5/3.0	6.2
	O/W	6	55.5	43	12.5	2.0/2.5	7.2
10%NaCl	W/O	401	63.5	50	13.5	3.0/4.0	7.4
	O/W	6	59.5	48	11.5	2.5/3.5	8.4

315 to 5 V and conductivity increases from 0 to 1.26 ms/cm. At an acid concentration of 4%, the emulsion is reversed to an O/W emulsion with a high conductivity and low ES value. However, the emulsion reversion experiences become unstable at an acid concentration of 2% when the pH is lower than 7. The oil-phase and water-phase resolutions increase simultaneously, suggesting demulsification. The hydrophilicity of RPEM increases because amine protonation occurs as the pH value decreases. This leads to the formation of an O/W emulsion. However, sedimentation is likely in the reversed O/W emulsion with a high water-phase resolution, which may be due to insufficient hydrophilic groups.

Figure 2 shows the variation in the ES value, conductivity, pH, and continuous phase resolution of the emulsion on the addition of the base. As the base concentration increases, the oil-phase resolution of the W/O emulsion stabilized by only RPEM is close to 0%. In addition, the ES value increases with the pH, indicating that the W/O emulsion stability increases under basic conditions.

We incorporated a secondary emulsifier (1% RZ-1) into the emulsion system to improve the emulsion reversion stability. **Figure 3** shows the variation in the ES value, conductivity, pH, and continuous phase resolution of the pH-responsive emulsion stabilized by the emulsifier on the addition of an acid.

The initial ES value of the W/O emulsion stabilized by 2% RPEM and 1% RZ-1 is 435 V, which meets the API standard (ES > 400 V). As the acid concentration increases to 3%, the ES value decreases to 5 V. In addition, the conductivity increases to 0.57 ms/cm, indicating the reversion to an O/W emulsion. Even though the water-phase resolution of O/W emulsions is 25%, demulsification does not occur at any acid concentration. Therefore, by decreasing pH value from 7 to 5, the protonation of amine groups renders the increasing hydrophilicity of emulsifiers, leading to stable reversion from a W/O emulsion to an O/W emulsion.

Figure 4 shows the variation in the ES value, conductivity, and continuous phase resolution of the pH-responsive emulsion stabilized



sample height for the W/O and (D) O/W emulsion-based drilling fluids.

TABLE 4 | Properties of reversible drilling fluid contaminated by drill cuttings.

Drill cuttings/%	Emulsion type	ES/V	AV/mPa⋅s	PV/mPa⋅s	YP/Pa	G _{10s} /G _{10min}	FL _{HTHP} /mL
0	W/O	422	51.5	40	11.5	3.0/4.0	4.8
	O/W	6	50.5	39	11.5	3.5/4.5	8.2
5	W/O	436	56.5	43	13.5	3.0/3.5	5.4
	O/W	6	58.5	46	12.5	3.5/4.0	6.2
10	W/O	461	60.5	46	14.5	3.5/4.5	7.2
	O/W	6	63.5	50	13.5	4.0/5.5	8.2

by the emulsifier on the addition of the base. The conductivity decreases as the base concentration increases. At a base concentration of 3%, the conductivity decreases to 0 ms/cm and the ES value increases to 100 V, suggesting the reversion from an O/W emulsion to a W/O emulsion. As the base concentration increases to 6%, the ES value of the W/O emulsion recovers back to 421 V and oil resolves minimally, indicating a stable W/O emulsion. Demulsification is not observed during the reversion from an O/W emulsion to a W/O emulsion.

Figures 4D,E show that the mean emulsion droplet sizes of the W/O and O/W emulsions before and after emulsion reversion are less than 21 μ m and uniformly distributed. It can be concluded that stable reversion from an O/W emulsion to a W/O emulsion and *vice versa* can be achieved by changing the pH values.

Drilling Fluid Performance High-Temperature Resistance

We examined the performance of W/O and O/W emulsion-based drilling fluids. Drilling fluid was prepared using the optimal formula:

TABLE 5 | Cleanup efficiency of reversible drilling fluid at different pH values.

pH value	Cleanup efficiency %	Water contact angle (°)		
7	47.2	116.5		
6	63.5	92.7		
5	86.3	56.4		
4	90.7	34.8		
3	95.1	23.6		
Surfactant flushing fluid	89.4	36.1		

2 wt% PREM+1 wt% RZ-1+1 wt% organoclay+3 wt% filtrate loss reducer. The drilling fluid density was adjusted to 1.5 g/cm^3 using barite. The drilling fluid was hot rolled for 16 h at various temperatures and then cooled to 65° C for performance testing according to the API standard. The results are presented in **Table 1**.

Based on the API standard for oil-based drilling fluids, on-site practice requires an ES value above 400 V to ensure emulsification



FIGURE 6 | (A) Viscometer cylinder covered by reversible drilling fluid before washing. (B) Cleanup efficiency for reversible drilling fluid washed by acid at pH = 5 and (C) pH = 3.

stability in a wellbore. **Table 1** shows that the ES value of the W/O emulsion-based drilling fluid is over 400 V even after hot rolling at temperatures of 100–150°C, indicating excellent emulsion stability even at a low oil-to-water ratio. Normally, low oil-to-water ratio usually makes it difficult to maintain high emulsion stability and suitable rheology (Oltedal et al., 2015). The rheological properties of the O/W emulsion-based drilling fluid after reversion are similar to those of the W/O emulsion-based drilling fluid, and there is no severe demulsification or barite sag during emulsion reversion. This suggests that the W/O and O/W emulsion-based drilling fluids exhibit excellent rheology and low filtration loss (less than 10 ml), which meets drilling engineering requirements.

Sag Stability

In high-density drilling fluids, weighting materials tend to settle because of gravity. This might result in a loss of circulation, drilling-pipe sticking, well-control difficulty, and cementing problems (Elkatatny, 2018). In reversible drilling fluids, weighting materials must be transferred between the oil and water phases during emulsion reversion, which might increase density variation. Therefore, the sag stability of reversible drilling fluids was examined at a density of 1.5 g/cm³. The drilling fluid density variations at the top and bottom sections were measured for W/O and O/W emulsion-based drilling fluids, and the results are shown in **Table 2**.

The SFs for both types of drilling fluids are less than 0.52, and the density variation is less than 0.05 g/cm³ (**Table 2**), confirming the prevention of barite sagging (Elkatatny, 2019). Weighting materials are well suspended in the system, and the density of the reversible drilling fluids is uniformly distributed, indicating excellent sedimentation ability.

A multiple light scattering analyzer is a nonintrusive instrument used to investigate destabilization mechanisms without dilution or stress (Mengual et al., 1999). The scattering and transmission rates for the W/O and O/W emulsion-based drilling fluids after hot rolling at 150°C for 16 h were measured, and the results are shown in **Figure 5**. **TABLE 6** | Reduction in oil content of drill cuttings generated by reversible drilling fluid.

pH value	Oil content o		
	drill cuttings/%		
Blank sample	35.5		
7	29.3		
6	19.5		
5	16.2		
4	3.2		
3	1.3		

The backscattering (BS) rates for the W/O and O/W emulsion drilling fluids overlap for 1 h. The mean values of the backscattering rate for the test sample height are almost the same for 1 h, suggesting that the emulsions are uniformly distributed. There is no creaming or sedimentation. These results show that the reversible drilling fluids with a density of 1.5 g/cm^3 achieve stable emulsion reversion even after hot rolling at 150° C for 16 h.

Contamination Test

Contamination, such as by saline water or drill cuttings, typically occurs when drilling into deep formations. Therefore, it is necessary to evaluate the performance of the reversible drilling fluids contaminated by various salts or drill cuttings. **Table 3** shows the properties of the drilling fluid contaminated by saline water.

The rheology of the contaminated reversible drilling fluid increases owing to the increased saline water ratio. More water droplets in the dispersed phase cause flow friction. On the other hand, the rheological properties for O/W emulsion are smaller than W/O emulsion when contaminated by saline water, as more continuous phase fraction for O/W emulsion. The ES values for the W/O emulsions remain above 400 V, indicating excellent salinity tolerance. The filtration loss for both W/O emulsion and O/W emulsion are less than 10 ml, which



meets the API standard. These results show that the properties of the reversible drilling fluid can be adjusted to maintain stability in wellbores even exposed to gypsum formation.

Table 4 shows the properties of the reversible drilling fluid contaminated with drill cuttings. Both W/O and O/W emulsion drilling fluid show excellent resistance to drill cuttings contamination. The AV, PV, and YP of the reversible drilling fluid increase with the amount of drill cuttings owing to the overload solid content that resists flow mobility. However, the rheological properties and filtration loss properties are within an acceptable range when the drilling fluid is contaminated with 10% cuttings.

Cleanup Efficiency

In the case of conventional oil-based drilling fluids, large amounts of surfactants and complicated cleaning procedures are required to remove the residual drilling fluid and filter cakes from wellbores and drilling pipes for high-quality cement bonding. Reversible drilling fluids are expected to improve the cleanup efficiency. To verify this, the cleanup efficiency of the as-prepared reversible drilling fluid was investigated using a rotational viscometer with flushing acids, and the results are shown in **Table 5**; **Figure 6**.

The cleanup efficiency of the reversible drilling fluid increases as the pH decreases, and the viscometer cylinder surface returns to the waterwet state. The introduction of H^+ ions into the system leads to the protonation of RPEM, which facilitates reversion to the O/W emulsion. Consequently, the residual O/W emulsion on the viscometer is easily washed by the inherently water-based flushing fluids used in the cementing procedure. Even though a high H^+ concentration results in a high efficiency, it is suggested to use flushing fluids at a pH of 4–5 owing to the corrosion risk of metals exposed to acids. The cleanup efficiency of the reversible drilling fluid using fluids at a pH of 4 is similar to that of using conventional surfactant flushing fluids.

Drilled Cutting Disposal

Oily drill cuttings that circulate on the surface cause severe environmental pollution. Therefore, it is necessary to investigate the possibility of reducing the oil content of the drill cuttings produced by reversible drilling fluids. The most common treatment technique used in the UK at present is indirect thermal desorption (Page et al., 2003). The oily drill cuttings produced by the reversible drilling fluid were washed with simple acids, and the resulting oil contents are shown in **Table 6**.

As shown in ; **Figure** 7, the oil content of the drill cuttings reduces from 35.5% to 1.3% when they are exposed to acids. The lower pH values help reverse oil-wet drill cuttings into water-wet drill cuttings. Then, the drill cuttings return to their original orange color and easily disperse in water. In contrast, a high oil content remains for cuttings that are immersed in neutral liquids. Therefore, a reversible drilling fluid is beneficial for drilled cutting that are washed with acids, which provides a promising method to dispose oily drilled cuttings in cost-effective ways.

CONCLUSION

A type of pH-responsive drilling fluid with a temperature resistance up to 150°C and a density of 1.5 g/cm³ was prepared for deep or ultra-deep wells. Stable reversion from a W/O emulsion to an O/W emulsion and vice versa was realized, which paved the way for practical reversion in the wellbore for different geological formation. The W/O and O/W emulsions exhibited small droplet sizes and uniform distributions. The W/O and O/W emulsion-based drilling fluids showed excellent resistance to coalescence or sedimentation on the basis of light backscattering and SFs. There was no demulsification of the emulsions or barite sedimentation during emulsion reversion. The W/O and O/W emulsion-based drilling fluids showed similar properties after hot rolling at 150°C for 16 h. The W/O emulsion exhibited excellent resistance to contamination by saline water and drill cuttings. The residual W/O emulsion-based drilling fluid was washed with acids at a high efficiency. The oil content of the drill cuttings generated by the reversible W/O emulsion-based drilling fluid was reduced using acids.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

Conceptualization, LL. Funding acquisition, DL, JJ; review and editing, LL. DZ, JJ; Investigation, LL, XW, DL.

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Conflict of Interest: Author DL is employed by the SINOPEC.

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