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Article

Stability mechanism and ultrasonic destabilization mechanism of polymer waste drilling fluid

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Abstract: Polymer waste drilling fluid has extremely high stability, and it is difficult to separate solid from liquid, which has become a key bottleneck problem restricting its resource recycling. This study aims to reveal the stability mechanism of polymer waste drilling fluid and explore the destabilization effect and mechanism of ultrasonic waste drilling fluid. Surface analysis techniques such as X-ray energy spectrum and infrared spectrum were used in combination with colloidal chemical methods to study the spatial molecular structure, stability mechanism, and ultrasonic destabilization mechanism of drilling fluid. The results show that the particles in the drilling fluid exist in two forms: uncoated particles and particles coated by polymers, forming a high molecular stable particle system. Among them, rock particles not coated by polymer follow the vacancy stability and Derjaguin-Landau-Verwey-Overbeek (DLVO) stability mechanism, and the weighting material coated by the polymer surface follows the space stability and DLVO stability mechanism. The results of ultrasonic destabilization experiments show that after ultrasonic treatment at 1000 W power for 5 min, coupled with the addition of 0.02% cationic polyacrylamide, the dehydration rate is as high as 81.0%, and the moisture content of the mud cake is as low as 29.3%, achieving an excellent solid-liquid separation effect. Ultrasound destabilizes polymer waste drilling fluid by destroying the long-chain structure of the polymer. This study provides theoretical support and research direction for the research and development of polymer waste drilling fluid destabilization technology.

Keywords: waste drilling fluid; polymer; stability; ultrasound; destabilization

1. Introduction

Drilling fluid is the circulating flushing medium in the borehole during the oil and gas drilling and production process. It can be divided into water-based [1], oil-based [2], synthetic-based drilling fluid [3], etc. After repeated use, drilling fluid becomes waste drilling fluid, which carries a large amount of rock particles, weighting materials, various polymer additives, heavy metals, and crude oil, which is extremely harmful to the environment [4,5]. According to the “2022 Ecology and Environment Bulletin of the People’s Republic of China”, the amount of waste drilling fluid discharged from offshore oil and gas platforms in China in 2022 was as high as 141,000 m^3, and the discharge from a single well was 200–800 m^3 [6]. In recent years, environmentally friendly polymer water drilling fluids have been widely used [7]. A large amount of lubricants, fluid loss reducers, anti-collapse agents, viscosity reducers, shale inhibitors, stabilizers, emulsifiers, plugging agents, and other functional additives are added during their preparation [8,9]. The solid content of the abandoned drilling fluid is as high as 30%–50% [10]; the organic matter content is as high as 26 $g \cdot L^{-1}$ [11]; and the zeta potential is as low as −38 mV [6]; the organic and inorganic components interact to form a highly stable system of non-Newtonian fluid state, which makes it difficult to separate the solid and liquid of the waste drilling fluid, seriously restricting the recycling of resources.

At present, the high stability mechanism of polymer waste drilling fluid is still imperfect. Previous studies often use the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory to explain the stability mechanism of polymer waste drilling fluid. For example, Omland [12] used the DLVO theory to explain the stability of non-Newtonian fluid drilling fluid; Liu et al. [13] used zeta potential as an indicator of waste drilling fluid stability based on the DLVO theory to guide the development of a hyperbranched flocculant technology. However, the DLVO theory ignores the steric hindrance caused by the polymer in the system [14]. Previous studies have found that in polymer-stabilized particle systems, the main factor for stability is the adsorbed polymer layer rather than the diffusion layer [14,15]. Therefore, it is difficult to explain the high stability of polymer waste drilling fluids using a single DLVO theory, and it is urgent to study its stability mechanism in depth to guide the development of efficient destabilization technology.

At present, the main method for treating waste drilling fluid is to use polyferric sulfate, but the high concentration of sulfate ions remaining in the liquid phase restricts the reuse of the filtrate, and alternative technologies are urgently needed. Many previous studies have shown that the ultrasonic method has a good destabilization effect on stable systems containing high solid phases [16–18]. Ultrasound can form cavities through the cavitation effect, and local high temperature and high pressure are generated at the moment of cavity rupture, and hydroxyl radicals are generated [19]. Hydroxyl radicals are highly oxidizing, with a redox potential of 2.8 V, which can effectively oxidize and degrade various components in the system and achieve destabilization [20]. Studies have shown that ultrasound has a good destabilization effect on high-solid municipal sludge systems [16,17]. Therefore, it is very likely that the use of ultrasound can achieve efficient destabilization of waste drilling fluid.

In view of the current situation that the stability mechanism of polymer waste drilling fluid and the ultrasonic destabilization mechanism are unclear, this study intends to use physical and chemical analysis combined with mathematical simulation to study the stability mechanism of polymer waste drilling fluid and further study the destabilization efficiency and mechanism of ultrasound on waste drilling fluid. This study is expected to solve the problem of solid-liquid separation of polymer waste drilling fluid and provide theoretical support and technical guarantees for clean production in oil fields.

2. Materials and methods

2.1. Source and properties of drilling fluid

The polymer waste drilling fluid comes from a drilling operation block in Bohai Oilfield. Its main components are polymers such as polyamine inhibitors, resin anti-collapse agents, alcohol amine coating agents, viscosity-enhancing and shear-enhancing agents, and synthetic ester lubricants, as well as particles such as rock particles and weighting materials and inorganic salts. Its density is 286.20 $g \cdot L^{-1}$ and its water content is 67.54%.

2.2. Methods for stability analysis

Zeta potential of the waste drilling fluid was analyzed by a nanoparticle size and zeta potential analyzer (ZS90, Malvern, UK). After freeze-drying, the samples were analyzed for chemical bonds and functional groups by a Fourier-transform infrared spectrometer (FTIR) (Nicoleti N10MX, Thermo Fisher Scientific, USA). The particle micromorphology and elemental composition were analyzed by a scanning electron microscope-energy dispersive spectrometer (Quattro, FEI, USA) for SEM-EDS analysis.

2.3. Research methods for ultrasonic destabilization mechanism

The ultrasonic generator is an ultrasonic cell disruptor (LC-JY16-11D, Shanghai Lichen Instrument Technology Co., Ltd.), which uses a $\varphi 6$ standard amplitude rod and an ultrasonic power adjustment range of 0–1000 W. Each time, 50 mL of waste drilling fluid was measured and poured into a 100 mL beaker and pretreated according to the set ultrasonic power and ultrasonic time. The organic flocculant is cationic polyacrylamide (CPAM), with a molecular weight of 8 million to 10 million and an ionicity of 30%–35%, and was prepared as a CPAM solution with a mass fraction of 1%. During flocculation, CPAM solution is added and slowly stirred for 1–2 min to allow the CPAM polymer chain to fully contact the particles to produce a flocculation effect.

The viscosity of the waste drilling fluid after ultrasonic destabilization was analyzed using a rheometer (ConTec-Viscometer 5, Fungilab, Spain), and the particle size distribution was analyzed using a laser particle size analyzer (Mastersizer 3000, Malvern, UK).

2.4. Analysis of solid-liquid separation effect

The treated waste drilling fluid was centrifuged at 5000 g for 5 min, and the volume of the supernatant was measured to calculate the dehydration rate. The mud cake after centrifugation was dried in an oven at 105 °C for 2 h according to the “Test Method for Sludge from Urban Wastewater Treatment Plants” (CJ/T 221-2005), and the moisture content was calculated.

3. Results and discussion

3.1. Stabilization mechanism of polymer waste drilling fluid

3.1.1. Stability analysis

The stability of polymer waste drilling fluid was studied. After centrifugation at 5000 g for 5 min, the waste drilling fluid did not undergo solid-liquid separation and remained in the initial viscous state; no flocculation occurred after adding CPAM to the waste drilling fluid. The results of zeta potential analysis showed that the zeta potential of the polymer waste drilling fluid was −31.7 mV, while the zeta potential after adding 0.02% CPAM was −30.9 mV, indicating that CPAM had little effect on the stability of the polymer waste drilling fluid. It is generally believed that when the zeta potential is higher than 30 mV or lower than −30 mV, the system is in a highly stable state [15], indicating that the polymer waste drilling fluid is a highly stable system.

3.1.2. Analysis of particle micromorphology and spatial structure

The solid content of polymer waste drilling fluid is as high as 32.46%, mainly containing weighting materials and rock particles; at the same time, it contains polymers with a cumulative proportion of more than 5%, mainly including polyamine inhibitors, resin anti-collapse agents, alcohol amine coating agents, viscosity enhancers and fluid loss reducers, synthetic ester lubricants, etc. The components in the waste drilling fluid were prepared into suspensions, and their zeta potentials were analyzed. It was found that the weighting materials were positively charged, while the rock particles and polymers were negatively charged (**Table 1**). Based on the charge characteristics, it is speculated that the rock particles in the solid phase of the drilling fluid may be repelled by the polymer through electrostatic repulsion to achieve high dispersion and stability, while the weighting material may adsorb the polymer through electrostatic attraction to form a stable structure of polymer-wrapped particles. The SEM-EDS technology was further used to conduct a two-dimensional element distribution analysis of the characteristic element Si in the rock particles and the characteristic element Ba in the weighting material (**Figure 1**). The results showed that the signal of Si is strong, while the signal of Ba is extremely weak, indicating that the surface of the rock debris particles is exposed and not wrapped, while the weighting material is wrapped by high molecular polymer. This result preliminarily verifies the above speculation.

Table 1. Zeta potential of formula components of polymer waste drilling fluid.

Formula components of polymer waste drilling fluid	**Zeta potential/mV**
Weighting materials	12.1
Rock particles	−11.3
Polyamine inhibitors	−21.6
Alcohol amine coating agents	−17.7
Resin anti-collapse agents	−9.6
Synthetic ester lubricants	−8.2
Viscosity enhancer and fluid loss reducer	−2.9

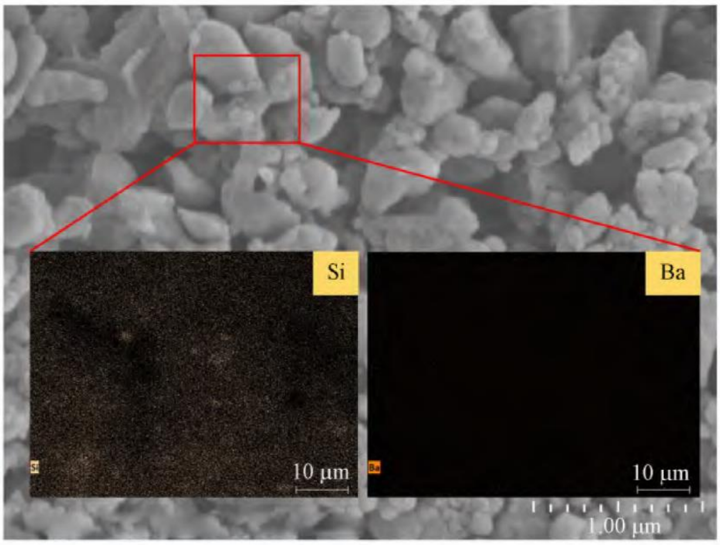

Figure 1. Two-dimensional element distribution analysis.

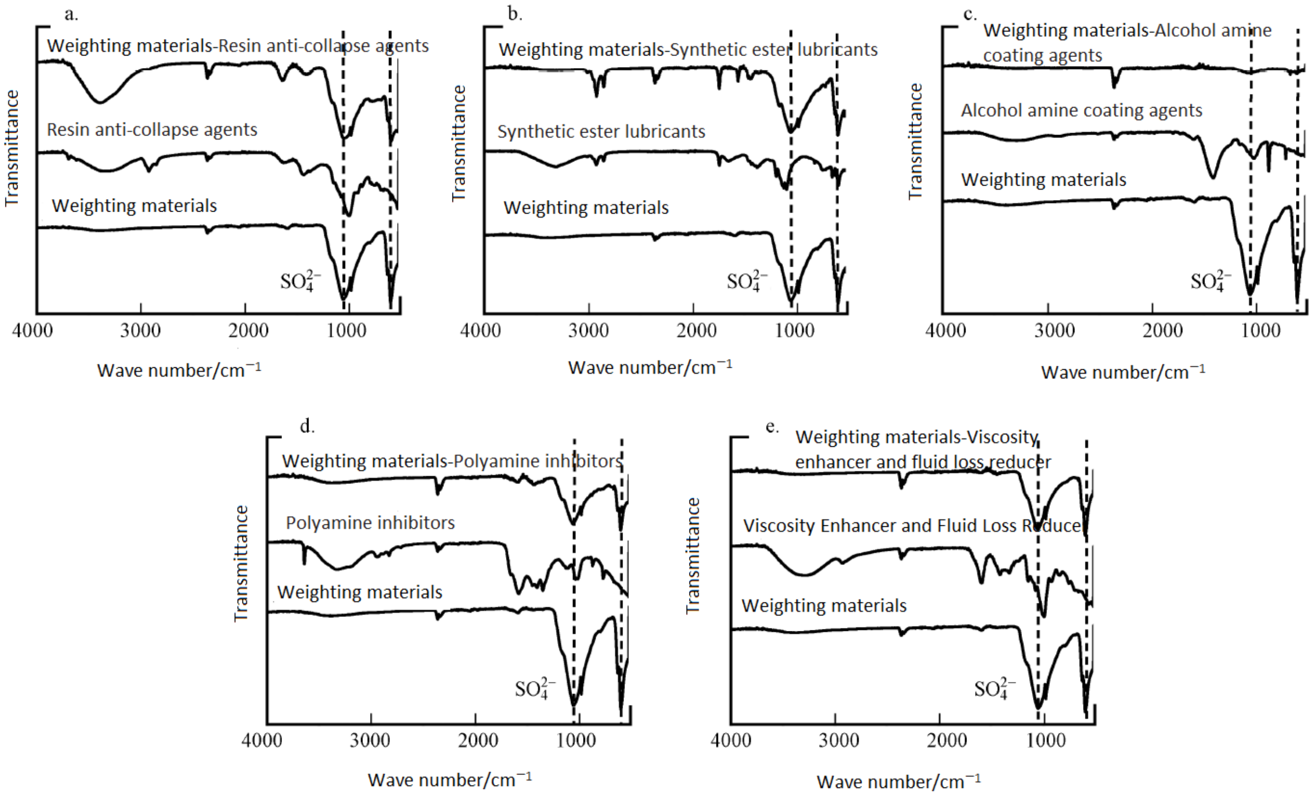

Figure 2. FTIR analysis of polymer components and their mixture with weighting materials.

In order to reveal the spatial bonding mode between polymers and weighting materials, the mixed samples of five drilling fluid polymers and weighting materials were freeze-dried and then analyzed by FTIR. As shown in **Figure 2**, the peaks of the weighting material at 1057 cm^{-1} and 981 cm^{-1} are the asymmetric and symmetric stretching vibration peaks of SO_4^{2-}, respectively, and the peaks at 630 cm^{-1} and 601 cm^{-1} are the bending vibration peaks of SO_4^{2-} [21]. Compared with the weighting materials, the peak intensity of SO_4^{2-} in the mixed samples of weighting materials-resin anti-collapse agents and weighting materials-synthetic ester lubricants is almost unchanged (**Figure 2a,b**), indicating that neither the resin anti-collapse agents nor the synthetic ester lubricants have wrapped the weighting materials. Compared with the weighting material samples, the peak intensity of SO_4^{2-} in the mixed samples of weighting materials-alcohol amine coating agents almost completely disappeared (**Figure 2c**), and the peak intensity of SO_4^{2-} in the mixed samples of weighting materials–polyamine inhibitors and weighting materials-viscosity enhancer and fluid loss reducer was weakened (**Figure 2d,e**), indicating that the polymers used to wrap the weighting materials are mainly alcohol amine coating agents, followed by polyamine inhibitors and viscosity-enhancing and shear-enhancing agents.

The above results show that the solid particles in the polymer waste drilling fluid exist in two ways. Among them, the rock particles exist in the polymer solution as individual particles mainly through electrostatic repulsion, while the weighting materials exist in the form of alcohol amine coating agents, polyamine inhibitors, and viscosity-enhancing and shear-enhancing agents mainly through electrostatic attraction.

3.1.3. Stabilization mechanism of polymer waste drilling fluid

Based on the above results, a stability model of polymer waste drilling fluid was constructed (**Figure 3**).

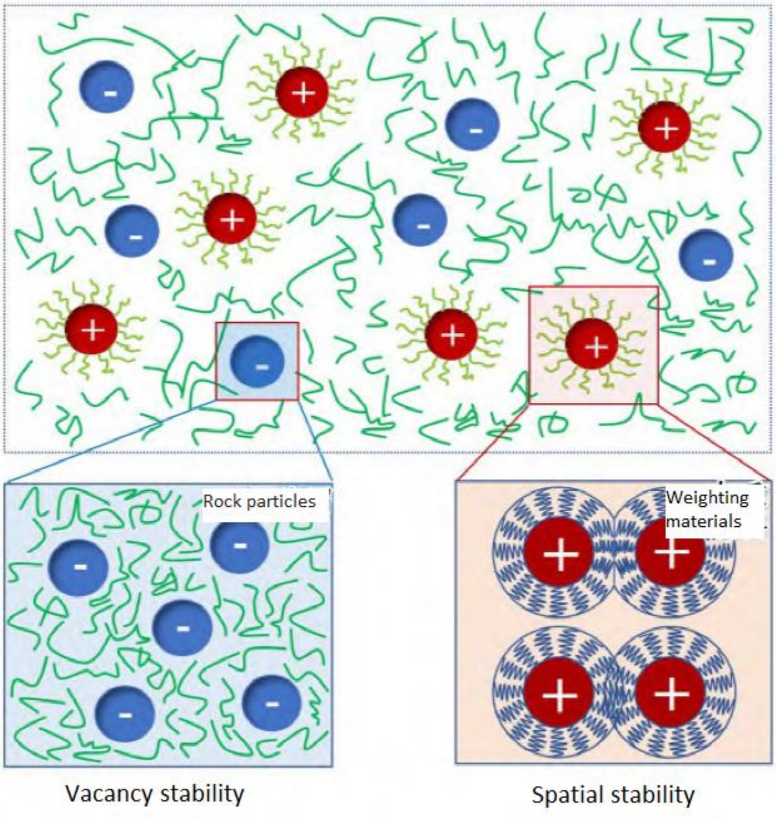

Figure 3. Stability mechanism of polymer waste drilling fluid.

1) DLVO and vacancy stability theory. It is difficult for molecular polymers to adsorb on the surface of rock particles, resulting in the polymer concentration on the particle surface being much lower than that in the liquid phase, forming a vacant surface adsorption layer. A large amount of polymers are dispersed in the liquid phase,

resulting in the dominance of repulsive potential energy, forming vacancy stability [14]. At the same time, there is still a double electrical layer on the surface of particles that are not adsorbed with high molecular polymers. Therefore, the stability of rock particles can be explained by DLVO combined with vacancy stability theory.

2) DLVO and steric stabilization theory. The weighting material particles and the polymer have opposite charges. Due to electrostatic attraction, a large amount of alcohol amine coating agent, viscosity enhancer and shear enhancer, and polyamine inhibitor are adsorbed on the surface of the weighting material particles, forming a tightly wrapped polymer layer. The presence of the polymer layer will reduce the Hamaker constant between the particles, thereby reducing the van der Waals attractive potential energy ΔG_V (Equation (1)); the particle surface still has a charge, and there is an electrostatic repulsive potential energy ΔG_E caused by the double electric layer (Equation (2)). The two interaction energies, ΔG_V and ΔG_E, can be described by the DLVO theory (Equation (3)). In addition, due to the repulsion between polymers, a steric repulsive potential energy ΔG_S is generated (Equation (6)), including two effects: part of the polymer on the surface of the particle repels and compresses the other, and the polymer layers do not overlap, resulting in a repulsive interaction energy $\Delta \mathrm{G}_{VR}$ caused by the volume restriction effect (Equation (4)); some polymers can attract each other, the high molecular polymers on the surface of the particles penetrate each other, and the polymer layers partially overlap, resulting in the osmotic pressure limitation effect caused by the osmotic energy ΔG_M (Equation (5)). Therefore, the total interaction energy ΔG between the weighted material particles (Equation (7)) is a comprehensive reflection of the repulsive energy ΔG_{VR}, the osmotic energy ΔG_M, the van der Waals attractive potential energy ΔG_V, and the electrostatic repulsive potential energy ΔG_E [15,22]. Therefore, the stability of weighting material particles in drilling fluid can be explained by DLVO combined with spatial stability theory. The weighting material particles adsorb and wrap the surface polymer, resulting in a decrease in van der Waals attraction, an increase in the spatial repulsion formed by the volume restriction effect and the osmotic pressure restriction effect, and the total interaction energy is dominated by repulsion, and the particles repel each other, thus forming a highly stable system. Compared with the electrostatic repulsion in the DLVO theory, the spatial repulsion in the spatial stabilization mechanism has a longer range and plays a major repulsive role [14].

$$\Delta G_{\mathrm{V}} = \frac{A}{12\pi H^2} \tag{1}$$

$$\Delta G_{\mathrm{E}} = \frac{64 n_0 kT}{\kappa^2} \pi a \gamma_0^2 e^{-\kappa H} \tag{2}$$

$$\Delta G_{\mathrm{DLVO}} = \Delta G_{\mathrm{V}} + \Delta G_{\mathrm{E}} \tag{3}$$

$$\Delta G_{\mathrm{VR}} = 4vkT\left(\frac{1-12H^2}{il^2-1}\right)\exp\left(-\frac{6H^2}{il^2}\right) \tag{4}$$

$$\Delta G_{\mathrm{M}} = \frac{4\sqrt{6}\pi^2}{27}(\alpha^2-1)kTv^2h\left(\frac{6H^2}{il^2-1}\right)\exp\left(-\frac{3H^2}{il^2}\right) \tag{5}$$

$$\Delta G_{\mathrm{S}} = 2\pi a \int_{H_0}^{\infty} (\Delta G_{\mathrm{VR}} + \Delta G_{\mathrm{M}}) \mathrm{d}H \tag{6}$$

$$\Delta G = \Delta G_{\mathrm{DLVO}} + \Delta G_{\mathrm{S}} \tag{7}$$

where v is the number of polymer tails or rings on the particle surface; l is the length of the chain; i is the number of chain links; H is the particle distance; a is the particle radius; n_0 is the number of particles per unit volume; γ_0 is the particle conductivity; κ is the double layer thickness; α is the polymer expansion coefficient; h is the root mean square distance between the polymer ends; A is the Hamaker constant; k is the Boltzmann constant; and T is the system Kelvin temperature.

In summary, the polymer waste drilling fluid is a polymer-stabilized particle system in which the rock particles follow the vacancy stability theory and DLVO theory, and the weighting material follows the spatial stability and combined DLVO theory. The two work together to form the extremely high stability of the waste drilling fluid. Therefore, destroying the long chain of the polymer is one of the ways to achieve efficient destabilization of polymer waste drilling fluid.

3.2. Ultrasonic destabilization effect and mechanism of polymer waste drilling fluid

3.2.1. Ultrasonic destabilization effect

The destabilization effect of ultrasonic treatment on polymer waste drilling fluids was investigated. After 5 min of ultrasonic treatment at 1000 W, the zeta potential increased from −31.7 mV to −15.5 mV, indicating that ultrasonic treatment has a good destabilization effect. However, after centrifugation, the drilling fluid dehydration rate was only 60.50%, and the mud cake moisture content was only 59.27%. Further analysis of the particle size distribution of the drilling fluid after ultrasonic treatment showed that the average particle size (d_{50}) decreased from 82.7 μm to 18.5 μm, indicating that while ultrasonic treatment destabilized the waste drilling fluid, it also dispersed the solid phase components, reducing particle size and consequently leading to poor solid-liquid separation efficiency. Therefore, flocculants were used to enhance the solid-liquid separation efficiency of the drilling fluid after ultrasonic destabilization. After adding 0.02% CPAM, the average particle size (d_{50}) in the system increased from 18.5 μm to 227 μm, the drilling fluid dehydration rate improved to 81.00%, and the mud cake moisture content reached 29.30%, indicating that the combination of ultrasonic destabilization and CPAM flocculation achieved efficient solid-liquid separation of waste drilling fluid.

The effects of ultrasonic power and ultrasonic time on the destabilization of the waste drilling fluid were further studied. The ultrasonic time was kept at 5 min to study the effect of ultrasonic power on the destabilization of the drilling fluid. With a power of 200 W, the supernatant was viscous and turbid after centrifugation (**Figure 4a**), the drilling fluid dehydration rate was 68.50%, the mud cake water content was 57.36% (**Figure 4c**), and the zeta potential was −25.9 mV (**Table 2**), indicating that the ultrasonic destabilization effect was poor; with a power of 1000 W, the viscosity and turbidity of the supernatant after centrifugation were greatly reduced (**Figure 4b**), the drilling fluid dehydration rate was 81.00%, the mud cake water content was 29.30%, and the zeta potential was −1.9 mV, indicating that the ultrasonic destabilization effect was good. When the ultrasonic power was fixed at 1000 W and the ultrasonic time was increased from 1 min to 5 min, the dehydration rate increased from 70.00% to 81.00%, the moisture content of the mud cake decreased from 55.23% to 29.30% (**Figure 4d**), and the zeta potential increased from −21.4 mV to −1.9 mV (**Table 2**). With the increase of ultrasonic power or ultrasonic time, the dehydration rate gradually increased, and the absolute values of the moisture content and zeta potential of the mud cake gradually decreased, indicating that increasing the ultrasonic power or ultrasonic time can enhance the destabilization effect of the drilling fluid.

When the ultrasonic energy (power × time) was 1.2×10^5 J (**Table 2**), the zeta potential after 2 min of 1000 W ultrasonic treatment was −8.0 mV, which was higher than the −10.7 mV after 5 min of treatment with 400 W ultrasound; the dehydration rate of the former is 77.50%, which is higher than the 70.00% of the latter; the moisture content of the mud cake of the former is 40.30%, which is lower than the 55.20% of the latter. Under other ultrasonic energies, similar rules are also shown (**Table 2**); that is, when the ultrasonic energy is the same, the high-power short-time ultrasonic treatment has a higher zeta potential, a higher dehydration rate, a lower mud cake moisture content, and a better destabilization effect than the low-power long-time ultrasonic treatment. Therefore, the ultrasonic power has a greater influence on the destabilization effect than the ultrasonic time.

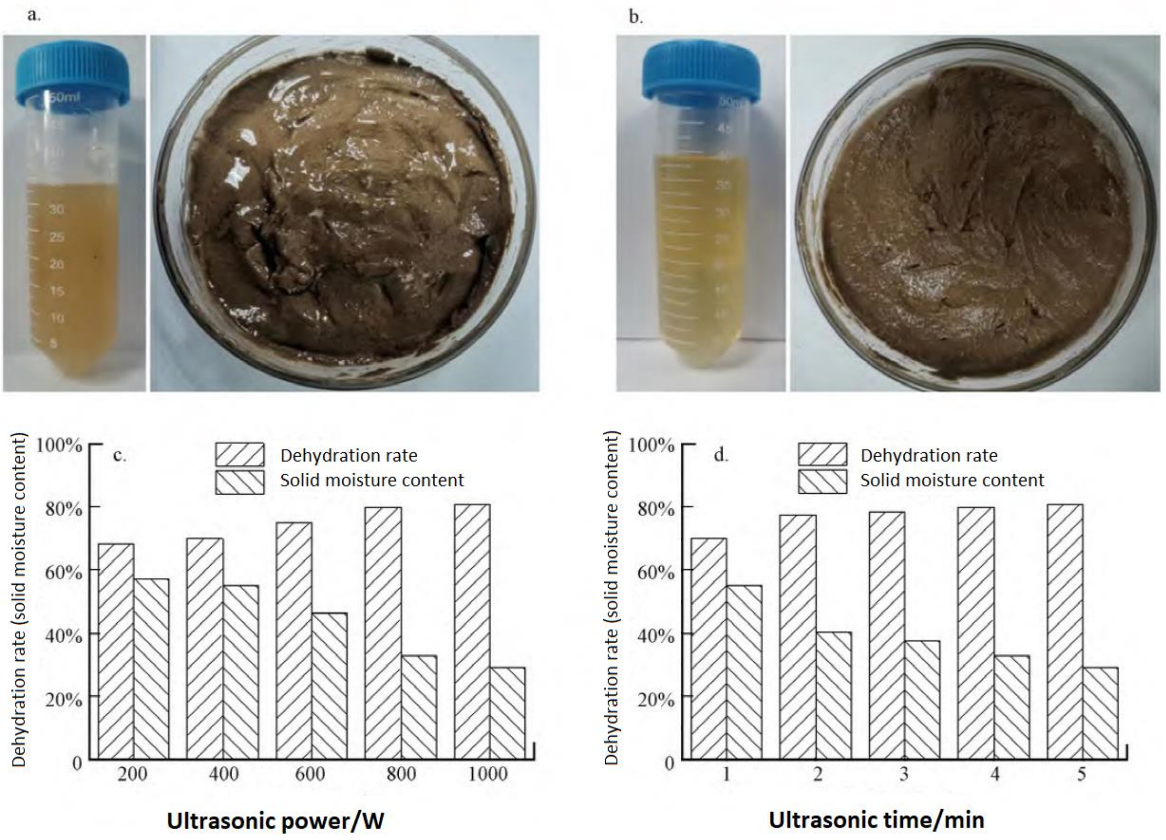

Figure 4. Ultrasonic destabilization efficiency of polymer waste drilling fluid. **(a)** 200 W power ultrasound for 5 min coupled with solid-liquid separation after adding CPAM flocculation; **(b)** solid-liquid separation effect after flocculation with CPAM added by coupling with 1000 W power ultrasound for 5 min; **(c)** the effect of ultrasonic power on the solid-liquid separation effect; **(d)** the effect of ultrasonic time on the solid-liquid separation effect.

Table 2. Zeta potential of polymer waste drilling fluid flocculated after ultrasonic treatment.

Ultrasonic power/W	**Ultrasonic time/min**	**Ultrasonic energy/10^4 J**	**Zeta potential/mV**
0	0	0	−31.7
200	5	6	−25.9
400	5	12	−10.7
600	5	18	−7.6
800	5	24	−3.2
1000	5	30	−1.9
1000	1	6	−21.4
1000	2	12	−8.0
1000	3	18	−4.3
1000	4	24	−2.8

3.2.2. Ultrasonic destabilization mechanism

The average molecular weight of polymers is positively correlated with their viscosity [15,23,24]. By measuring the change in system viscosity, the change in the molecular weight of polymers can be evaluated. The viscosity change of polymer waste drilling fluid before and after ultrasonic treatment is shown in **Figure 5**. The viscosity is 76.63 Pa·s without ultrasonic treatment. When the ultrasonic time is fixed at 5 min and the ultrasonic power increases from 200 W to 1000 W, the viscosity decreases from 48.52 Pa·s to 0.17 Pa·s (**Figure 5a**), which is 36.68%–99.78% lower than that without ultrasonic treatment; when the ultrasonic power is fixed at 1000 W and the ultrasonic time increases from 1 min to 5 min, the viscosity decreases from 25.54 Pa·s to 0.17 Pa·s (**Figure 5b**), which is 66.67%–99.78% lower than that without ultrasound. The above shows that ultrasound treatment destroys the long-chain structure of polymers, resulting in a decrease in polymer molecular weight, thereby reducing the viscosity of the system.

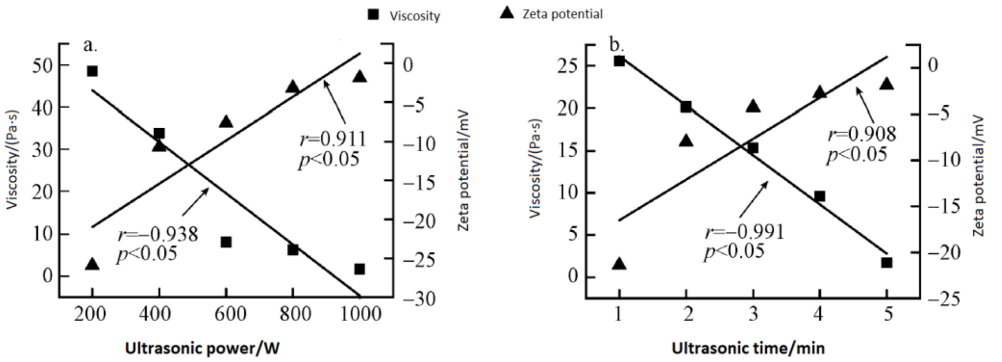

Figure 5. Relationship of viscosity and zeta potential to ultrasonic parameters. **(a)** Ultrasonic power; **(b)** ultrasonic time.

There is a significant negative correlation between ultrasonic power/time and system viscosity (**Figure 5a,b**), with correlation coefficients of −0.938 and −0.991, respectively; there is a significant positive correlation between ultrasonic power/time and system zeta potential (**Figure 5a,b**), with correlation coefficients of 0.911 and 0.908, respectively. This shows that increasing ultrasonic power and ultrasonic time can enhance the ultrasonic destabilization effect, and ultrasonic power is the main factor affecting the efficiency of ultrasonic destabilization. Ultrasound reduces the molecular weight by destroying the long chain of polymers, weakens the spatial resistance caused by polymers, and also reduces the zeta potential of the system, making the waste drilling fluid system efficiently destabilized.

4. Conclusions

1) Polymer waste drilling fluid is a polymer-stabilized particle system in which the rock particles not coated by the polymer follow the vacancy stability and DLVO stability mechanisms, and the weighting material coated by the polymer surface follows the space stability and DLVO stability mechanisms.

2) Ultrasound can greatly reduce the molecular weight of polymers by destroying their long-chain structures, thereby achieving efficient destabilization of drilling fluids. After ultrasonic destabilization, the use of an appropriate number of organic flocculants can significantly increase the particle size of the system, thereby improving the solid-liquid separation efficiency.

3) Increasing the ultrasonic power and ultrasonic time can enhance the ultrasonic destabilization effect, among which the ultrasonic power is the main factor affecting the ultrasonic destabilization efficiency. The destabilization effect of high-power short-time ultrasonic treatment is better than that of low-power long-time ultrasonic treatment.

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