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The Development and Evaluation of Novel Self-Degrading Loss-Circulation Material for Ultra-Deepwater Drilling in South China Sea

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Abstract: Focusing on the problem of drilling fluid loss circulation due to fractured granite formation in ultra-deepwater drilling in the research area, a novel self-degrading loss-circulation material is developed with polymer resin as the core material, according to the principles of loss-circulation prevention and reservoir and environmental protection. The relevant properties of the novel self-degrading loss-circulation material are evaluated by using tests or experiments. The pressure-bearing properties are evaluated by using a sealing capability test; the self-degrading properties are evaluated by using a self-degrading performance test; the pressure-bearing capability and reservoir protection properties are tested by measuring pressure sealing and gas permeability. The results of the tests and experiments show that the grinding rate of the loss-circulation material is less than 6% under 25 MPa, indicating that the novel loss-circulation material is of high compressive strength. The degradation process of the novel self-degrading loss-circulation material can be accelerated by increasing temperature and pH, and the degradation of the self-degradable polymer composite can be accomplished within 5 days under 95 °C and pH = 14. After self-degrading, the permeability recovery values of the highly permeable reservoir and the fractured reservoir are more than 96.9% and 99.15%, respectively, which indicates outstanding reservoir protection capability. Therefore, the novel self-degrading loss-circulation material has excellent temporary plugging and reservoir protection performance and can be used for plugging while drilling in ultra-deepwater drilling.

Keywords: ultra-deepwater; fractured granite formation; self-degrading loss-circulation material; South China Sea; reservoir protection

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1. Introduction

The deepwater area in the South China Sea is rich in oil and gas resources. A region with water depths of more than 500 m is called a deepwater zone, and those with water depths of more than 1500 m are called ultra-deepwater zones. In order to ensure the security of China's domestic oil and gas supply and solve the contradiction between supply and demand, China's oil and gas resources exploration and development has been steadily moving toward deepwater and ultra-deepwater oil and gas in recent years. The ultra-deepwater oil and gas resources in the Yongle block in the western South China Sea are rich in hydrocarbon reserves, with good prospects for exploration and development, and have large development potential, which is a key block for increasing reserves [1]. However, the pre-tertiary oil reservoirs in this block are granite weathering crust-fractured reservoirs, which is a metamorphic rock. Therefore, in the process of loss-circulation control while drilling, different types and sizes of solid-phase particles in drilling fluids can easily invade fractures around the borehole and cause contamination and damage to the reservoir [2–4]. Even with subsequent flushing, drainage and acidification, only a small portion of invaded drilling fluids and solid-phase particles can be removed, and the majority will remain in the reservoir pore space, causing serious contamination and damage to the reservoir, resulting in a significant reduction in oil and gas well production and making it difficult to achieve the goal of the efficient production of ultra-deepwater oil and gas [5,6].

In recent years, scholars have focused on oil and gas reservoir protection during the drilling process [7–10]. Current reservoir protection methods and unplugging techniques are mainly based on different plugging and reservoir conditions to provide a scientific and reasonable selection of pre-shielding temporary plugging techniques, post-acidification unplugging and biological enzyme unplugging techniques [11–14]. However, chemical, physical, microbial and compound plugging removal techniques are still required to deal with formation blockage. These unplugging processes are complicated, and if the unplugging measures are not properly handled, upcoming chemical and mechanical blockages will occur, which will reduce the permeability of the formation [15] and further increase the time and cost of ultra-deepwater operations. In the past two years, quite a few research works on self-degradation have been carried out. In 2018, Song Jifeng et al. [16] developed a self-degrading temporary plugging fluid system for the problem of sensitivity to water and water block caused by workover loss circulation in the Weizhou RRX oilfield in the South China Sea, which can achieve a 6-day self-degradation rate of more than 50% without broken gel. In 2020, Ye Lian et al. [17] tested a novel environmentally friendly self-degrading loss-circulation material for the problems of common temporary plugging materials not being able to self-degrade and insufficient pressure bearing in the plugged reservoir, of which the self-degradation rate can increase with increasing temperature, and the acidic and alkaline environments can promote its self-degradation effect. In 2021, Zhang Hao et al. [18] prepared a novel acid-soluble cured plugging material in order to solve the problem of severe loss-circulation control; in 2021, Song Zhaohui et al. [19] developed a reservoir protectant by grafting a partially crosslinking polymer on the surface of heavy calcium-carbonate-type rigid particles, with better acid solubility for reservoir properties and damage mechanisms in lowly permeable gas reservoirs in western Sichuan.

At present, the materials for oil and gas reservoir protection in the drilling process are mainly acidified materials, oil-soluble materials and biodegradable materials; however, all these materials require external auxiliary conditions to be unplugged and do not have fine self-degrading characteristics [20–23]. Therefore, in view of the contradiction that it is difficult to reconcile loss-circulation prevention and reservoir protection in current reservoir drilling processes, it is necessary to research and develop a novel plugging material that can perform self-unplugging, which can be a key technology to achieve oil and gas reservoir protection. Based on this, it is necessary to develop a self-degrading loss-circulation control material that can degrade chemically and generate small-molecule products under certain temperature and pressure conditions, within a certain time frame, so that it can be completely degraded [24–28]. This novel intelligent functional material can be used to seal reservoir fractures under pressure during drilling, and can also degrade and unplug itself in a reservoir environment at the later stage of completion.

By systematically exploring the self-degradation of the material and the characteristics of pressure sealing and self-unplugging, we optimized drilling fluid for ultra-deepwater plugging and developed a set of ultra-deepwater self-degrading plugging drilling fluid systems, which is expected to simultaneously solve the problem of loss-circulation and reservoir protection in fractured granite formations.

2. Preparation of Self-Degrading Loss-Circulation Material and Evaluation of Degradation Performance

2.1. Materials and Instruments

(1) Materials

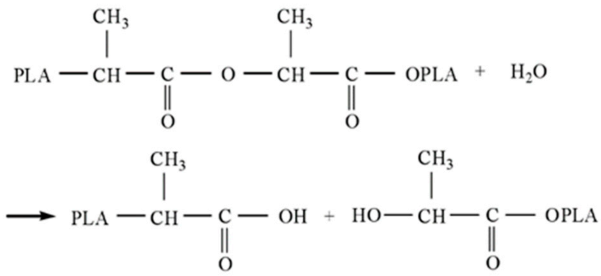
Polymer resin particles, polylactic acid, organic complexing accelerator, inorganic sodium salt, alkali hydroxide, triazine melamine organic matters, sea water, clean water.

(2) Instruments

Reaction apparatus, self-made crack plugging simulation test device, electric vacuum drying oven.

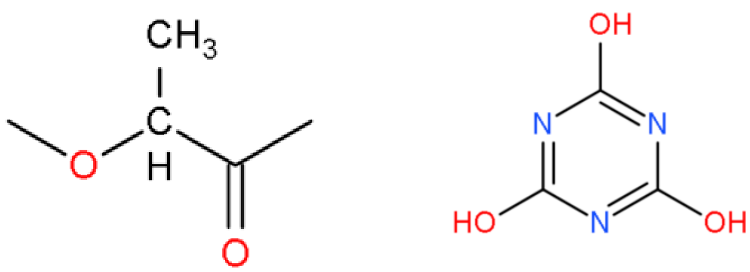
2.2. Design Principle and Preparation Method of Self-Degrading Loss-Circulation Material

Polylactic acid (PLA) is a biodegradable aliphatic polyester with lactic acid as a monomer, which boasts characteristics of non-toxicity, non-irritation, high strength, easy processing, excellent biocompatibility, biodegradable absorption and non-environmental pollution. Polylactic acid contains ester bonds that are sensitive to water and heat. In high-temperature, aerobic and humid environments, ester bonds are easily broken and hydrolyzed. The degradation equation of polylactic acid is shown in Formula (1):



(1)

However, the current biodegradation rate of PLA is slow and cannot meet the requirements of self-degradation after the drilling process. In response to this problem, thermally dissolved polymers and alkaline substances are introduced into PLA-like materials and complexed by organic complexation promoters, and triazine mimics organic substances to obtain a solid loss-circulation material with high strength. Under certain temperature conditions, thermally dissolved polymers, alkaline substances, etc., dissolve in advance, releasing active small molecules that are conducive to PLA degradation and can promote PLA degradation reactions, thus realizing high-efficiency degradation of PLA-like materials and achieving the goal of self-degradation. Among them, hot-melt polymer resins contain carbonyl groups and other structures, as shown in Figure 1a, and triazine mimics organic substances, with the structural formula shown in Figure 1b.



(**a**) (**b**)

Figure 1. Molecular structure of self-degrading loss-circulation material. (**a**) Molecular formula of polymer; (**b**) triazine melamine organic compounds.

Polymer resin particles are added into a reaction apparatus, heated to 175 °C using oil; then, polylactic acid, organic complexing accelerator, inorganic sodium salt and alkali hydroxide are added and stirred for 30–50 min; a certain amount of triazine melamine organic matters are added, then reacted for 10–15 min under stirring conditions; heating and stirring are stopped, the reactant is cooled down to obtain a solid product; the prepared solid product is crushed into solid particles with different particle sizes; thus, the self-degrading loss-circulation material is prepared.

A total of 10 g of self-degrading loss-circulation material and calcium carbonate particles with a particle size of 2.8–3.5 mm is weighed, placed in a pressure capsule, slowly pressurized to 25 MPa and then pressure is maintained for 5 min. After pressure relief, the materials are screened with a 2.8 mm screen grid, and the mass ratio of the remaining materials to raw materials and the crushing rate are calculated. The experimental results show that under 25 MPa, the crushing rate of the loss-circulation material is less than 6%, and it has great compressive strength. Moreover, its density is lower than 1.30 g/cm^3, and because the density of the drilling fluid is usually higher than 1.3 g/cm^3, it is, therefore, prone to suspend cuttings in drilling fluid. This was proven by corresponding experiments.

2.3. Experiment of Degradation Performance of Self-Degrading Loss-Circulation Material

The main components of the self-degrading loss-circulation material are polymer, organic weak acid molecules and inorganic small molecules, which can be automatically degraded into small molecules. Certain amounts of plugging materials are weighed and placed in water with different temperatures and pH levels (clean water or simulated brine; the formula of simulated brine is clean water + 2.67% NaCl + 0.23% $MgCl_2$ + 0.32% $MgSO_4$ + 0.12% $CaCl_2$), and self-degradation is observed. Then, it is washed with distilled water and dried in a vacuum oven at 60 °C. The quality of soaked particles is measured, and their degradation rate is calculated. The degradation rate is an important index to measure the degradation characteristics of degradable polymer materials. The calculation method is as follows:

$$S = (W_1 - W_2)/W_1 \times 100\% \quad (2)$$

where S is the degradation rate, %; W_1 is the quality of the plugging material before reaction, g; and W_2 is the mass of the remaining plugging material, g.

3. Evaluation of Self-Degrading Loss-Circulation Material Performance in Drilling Fluid

3.1. Thermal Stability

According to the evaluation using a thermogravimetric analyzer (TGA), the thermal decomposition process of the self-degrading loss-circulation material was divided into three stages: the first stage occurred when the temperature was 25~300 °C, and in this stage, water and other volatile substances of polymer were lost; the second stage occurred when the temperature was 300~382 °C, the thermal weight loss rate reached 90%, and the mass of polymer decreased rapidly in this stage, which was the decomposition and precipitation process of organic components and the main thermal decomposition stage; and the third stage occurred when the temperature was above 382 °C, and the final residue was of partial carbonization and other ash. The experiment showed that the thermal decomposition reaction would not occur when the temperature was below 300 °C. The downhole ambient temperature, in which the self-degrading loss-circulation material was used, was about 100 °C, so the self-degrading loss-circulation material had good thermal stability.

3.2. Evaluation of Degradation Performance of Self-Degrading Loss-Circulation Material

The experimental results of the degradation performance of the loss-circulation material at different temperatures, particle sizes, pH and additions are shown in Figure 2. Figure 2a shows the influence of temperature and water quality on the degradation rate. It can be concluded that under different temperatures and water quality, the self-degrading loss-circulation material can effectively self-degrade. In a clean water environment at 70 °C, the degradation rate of the loss-circulation material was very slow in the first 6 days, and there was no basic change, which can play a significant role in loss-circulation prevention and plugging. After 6 days, the physical and chemical degradation reaction of the loss-circulation material continued, and the degradation rate increased. As the degradation reaction gradually penetrated from the surface of the loss-circulation material, the contact area became larger, and more substances participated in the degradation reaction, which synergistically accelerated the degradation rate of the loss-circulation material. The loss-circulation material basically completed degradation in 10 days, and the self-degradation rate was close to 100%. In clean water at 95 °C, the loss-circulation material began to degrade rapidly after 5 days and basically completed degradation after 9 days. However, in a brine environment, the degradation time of the loss-circulation material had little change compared with that in clean water, and the degradation rate increased slightly, which indicates that a brine environment does not affect the degradation performance of the self-degrading loss-circulation material. This is because univalent and bivalent cations in brine neither affect alkaline substances in the loss-circulation material to accelerate the decomposition of polylactic acid nor inhibit the decomposition of polylactic acid itself. Despite this, cations will combine with carboxylic acid groups in decomposition products, thus slightly accelerating hydrolysis.

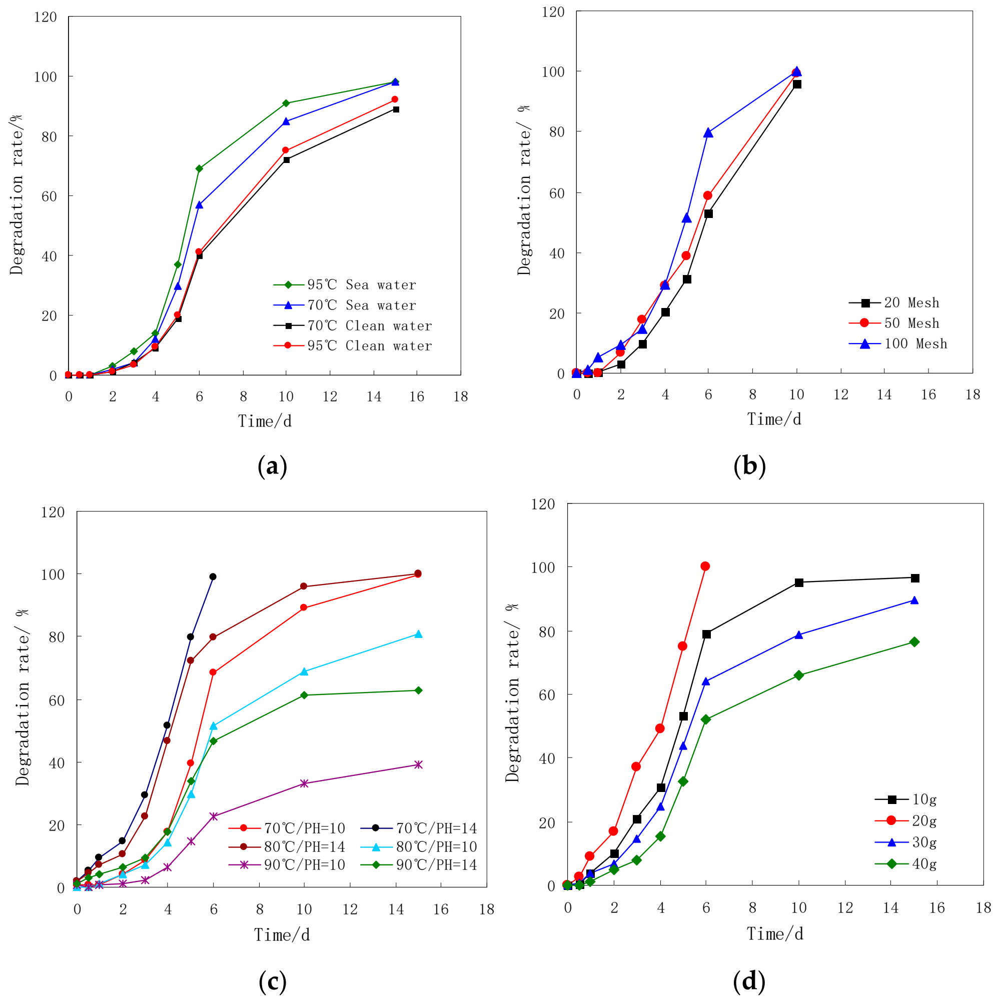


Figure 2. Degradation of loss-circulation material varies with time under different conditions. (**a**) Clean water/sea water, 70 °C/95 °C; (**b**) 20 mesh, 50 mesh, 100 mesh; (**c**) 70 °C, 80 °C, 90 °C and pH = 10, 14; (**d**) Addition of loss-circulation material. Note: in the experiment, except for the test items, other indicators are clean water, 80 °C, pH = 10; loss-circulation material is 20 mesh, and addition is 10 g.

Figure 2b shows the effect of particle size on the self-degradation rate. In the experiment, the selected particle sizes are 20 mesh, 50 mesh and 100 mesh; this is related to the common fracture width of granite reservoirs in the study area. The larger the particle size of the self-degrading loss-circulation material is, the faster the degradation rate is. This is because, with an increase in the mesh number, the specific surface area of self-degrading loss-circulation material increases, the contact area with water increases and the degradation of materials is accelerated. Therefore, there is a positive correlation between the particle size and the degradation rate of self-degrading loss-circulation material particles.

Figure 2c shows the effect of pH on the self-degradation rate. Under different pH levels (pH = 14 and pH = 10), it can be observed that the higher the pH is, the faster the degradation rate of self-degrading loss-circulation material is. This is because the ester bond in polylactic acid materials is more likely to break in a more alkaline environment, resulting in decomposition. In addition, this is also due to carboxylic acid products generated by the hydrolysis of the self-degrading loss-circulation material, which promotes a hydrolysis reaction to a positive reaction. When pH increases, the self-degradation of loss-circulation material will accelerate; therefore, pH is another main factor affecting the self-degradation of loss-circulation material particles. Therefore, under the dual action of temperature and pH, the degradation rate of the self-unplugging loss-circulation material will be further affected, and the degradation rate can be further accelerated at higher temperatures and an appropriate pH.

Figure 2d shows the effect of the addition of loss-circulation material on the self-degradation rate. After adding the 2.5% (10 g) self-degrading loss-circulation material, the degradation rate was low at the early stage and accelerated after a period of time, which follows the rule of the self-degrading loss-circulation material. After adding the 5% (20 g) self-degrading loss-circulation material, the degradation rate accelerated. This is because the amount of acid produced at the beginning of decomposition is enough to lower the pH of the solution and decompose acid-soluble materials. However, with the increased addition of the loss-circulation material, the degradation rate of the loss-circulation material decreased when it reached 7.5% (30 g) or 10% (40 g). This may be due to the fact that the decomposed substance is saturated in the solution, which inhibits the decomposition, or the area of the self-degrading loss-circulation material and liquid-phase release remains unchanged when the addition is increased, which leads to slow degradation.

In Reference [3], the effects of temperature, PH and electrolyte on the degradation rate were studied, and the research results were similar to those of this paper. However, the effects of particle size and the addition of loss-circulation material on the degradation rate were not studied in Reference [3].

3.3. Loss-Circulation Control Performance While Drilling

Through the micro-fracture plugging test of drilling fluid, the plugging performance of self-degrading loss-circulation material was analyzed, which provides a reference for optimizing the ultra-deepwater plugging drilling fluid system. With the help of a fracture plugging simulation apparatus, cores with different fracture widths were selected. The selection of the fracture width refers to logging the data of the granite reservoir in the Yongle block in the South China Sea (Table 1). In this paper, the plugging performance of the self-degrading loss-circulation material while drilling was mainly investigated, so the fracture widths were set to 0.2 mm, 0.4 mm, 0.6 mm and 0.8 mm, and the plugging performance of the loss-circulation material for a 1.0 mm fracture was tested.

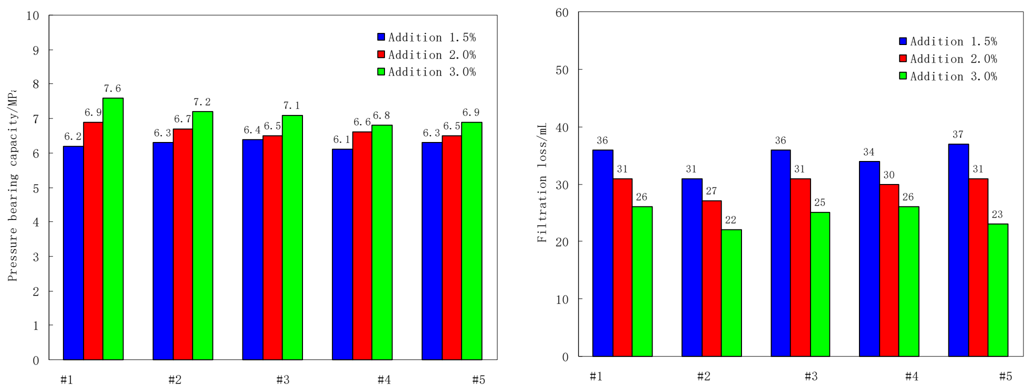
Table 1. Fracture width and loss-circulation analysis of granite reservoir in Yongle block, South China Sea.

Fracture Width/mm	Loss-Circulation Status	Solution
0~0.2	No loss	Take loss-circulation prevention measures while drilling
0.2~0.8	Slight loss	Take loss-circulation prevention measures while drilling
0.8~1.0	Small amount of loss	Stop drilling to take loss-circulation prevention measures
1.0~4.0	Large amount of loss	
>4.0	Severe loss	

According to the different fracture widths, the particle size distribution combination of the self-degrading loss-circulation material was preliminarily optimized, as shown in Table 2. The plugging performance was tested in the following ways: the cores of different fracture widths were placed in a high-temperature and high-pressure pressure plugging apparatus; the plugging material was added into the drilling fluid to prepare the plugging drilling fluid; the drilling fluid was added to the apparatus; the pressurizing valve and the liquid outlet at the lower end were opened; a measuring cylinder was placed directly below the liquid outlet and slowly pressurized and stabilized for a period of time (or broken through); then, the pressure-bearing capacity was recorded, the volume of filtration loss was measured and the plugging capacity of micro-fractures was evaluated, as shown in Figure 3. In this paper, the formula of the ultra-deepwater drilling fluid is as follows: fresh water + 1.0% filtration reducer PF-FLOTROL + 0.3% PF-PLUS + 0.3% PF-XC + 0.7% lubricant PF-HLUB + 3.0% inhibitor PF-UHIB + 2.5% calcium carbonate PF-EZCARB + 5% KCl + 15% NaCl. The formula of the ultra-deepwater plugging drilling fluid is as follows: base slurry + 1.5~3.0% plugging material.

Table 2. Particle size distribution of plugging materials combination for different widths.

Loss-Circulation Material	#1	#2	#3	#4	#5
Fracture width	0.2 mm	0.4 mm	0.6 mm	0.8 mm	1.0 mm
Particle size distribution	0.061~0.10 mm	0.061~0.30 mm	0.074~0.50 mm	0.089~0.71 mm	0.089~0.84 mm



(**a**) (**b**)

Figure 3. Plugging performance of self-degrading loss-circulation material. (**a**) Pressure-bearing capacity; (**b**) filtration loss.

Figure 3 shows the influence of the self-degradable plugging material on the plugging performance of the ultra-deepwater drilling fluid. From the lab results, it can be concluded that the self-degrading loss-circulation material has a fine plugging ability for plugging material combinations of different fracture widths, and with an increase in the plugging material combination, the pressure-bearing capacity of the plugging layer formed by ultra-deepwater drilling fluid gradually increases. For cores with a fracture width of 0.2 mm, after plugging with 3% drilling fluid, the pressure can reach 7.6 MPa, and the loss-circulation volume is 26 mL, which is relatively low. For cores with a fracture width of 1.0 mm, the pressure can still reach 6.9 MPa after plugging. This is because the plugging materials with different particle sizes can play a synergistic role, improve the deformation and particle size degradation rate of the plugging material under pressure and form a pressure-bearing plugging layer supported by plugging materials. The self-degrading loss-circulation material has high strength and a strong surface friction coefficient, which improves its retention and bridging function. The aforementioned actions cooperate with each other and greatly improve plugging stability.

3.4. Compatibility

GB/t16783.1-2014 "Field testing of drilling fluids—Part 1: Water-based fluids" and GB/T29170-2012 "Petroleum and natural gas industries-Drilling fluids-laboratory testing" tested the filtration loss and rheological properties of ultra-deepwater drilling fluids with different material additions and analyzed the compatibility between plugging materials and drilling fluids. Figure 4 shows the API filtration loss of the drilling fluid under different plugging systems and different additions. It can be seen that for different plugging materials, with an increase in additions, the filtration loss of the ultra-deepwater drilling fluid has little change, which indicates that plugging materials have little influence on the filtration loss of drilling fluid.

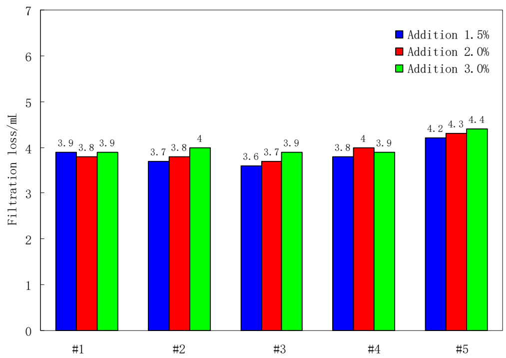


Figure 4. Influence of self-degrading loss-circulation material on filtration loss of current ultra-deepwater drilling fluid.

Figure 5 shows the compatible rheology of the ultra-deepwater drilling fluid at different temperatures. It can be concluded that the ultra-deepwater drilling fluid has fine rheology at different temperatures (4~80 °C). At low temperatures (4 °C), the funnel viscosity of drilling fluid increased slightly, but it still maintained fine fluidity. After adding plugging materials, the rheology of ultra-deepwater drilling fluid changed little, the funnel viscosity and apparent viscosity remained low and the rheology was stable. The initial gel strength and final gel strength increased slightly, but the changes were negligible. After being hot-rolled at 80 °C, the viscosity and yield point of the drilling fluid decreased slightly, but the yield point was 6.0~8.0 Pa. There was no phase separation after 24 h of being hot-rolled, which indicates that the self-degrading plugging material has little influence on the rheological properties of drilling at ultra-deepwater temperatures.

3.5. Removal Performance of Filter Cake

The #2 loss-circulation material was selected to prepare the ultra-deepwater plugging drilling fluid system, and the formula is ultra-deepwater drilling fluid + 3% loss-circulation material. According to the standard GB/T 16783.1-2014 "Field testing of drilling fluids—Part 1: Water-based drilling fluids" and GB/T29170-2012 "Petroleum and natural gas industries-Drilling fluids-laboratory testing", a drilling fluid filter cake was prepared by using a filtration loss meter. The filter cake was soaked in clean water at 80 °C and its degradation was observed, as shown in Figure 6. It can be seen that the filter cake formed by the drilling fluid system can self-degrade at 80 °C, and the thickness of the filter cake is obviously reduced after 7 days, from 0.7 mm in one day to 0.3 mm, which shows that the drilling fluid filter cake formed by the self-degrading loss-circulation material still has an excellent self-degrading ability, which can significantly reduce damage to the reservoir.

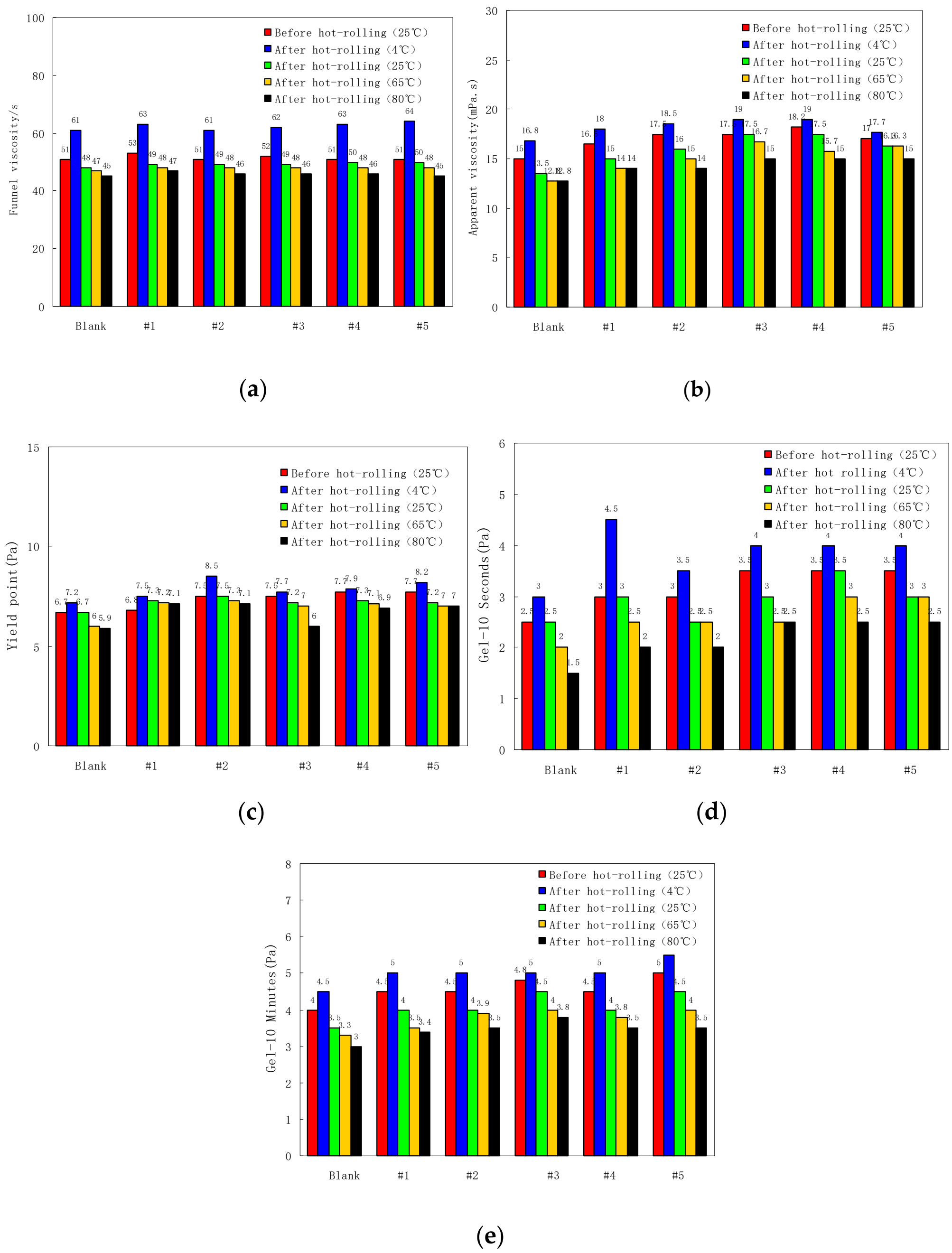


Figure 5. Compatibility of self-degrading loss-circulation material with ultra-deepwater drilling fluid. (**a**) Funnel viscosity; (**b**) apparent viscosity; (**c**) yield point; (**d**) Gel-10 s; (**e**) Gel-10 min.

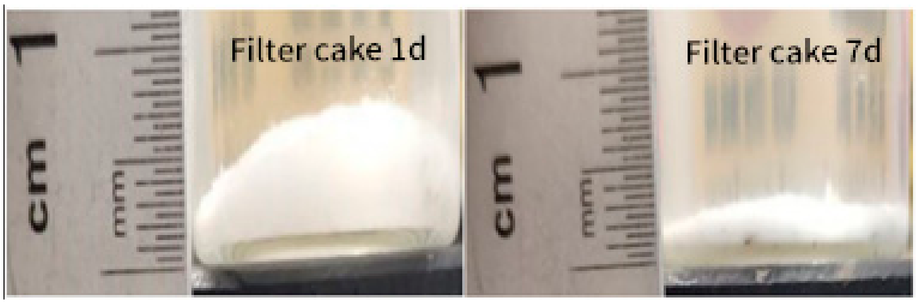


Figure 6. Self-degrading performance of ultra-deepwater drilling fluid filter cake.

3.6. Performance of Reservoir Protection

According to the People's Republic of China (PRC) petroleum and natural gas industry standard SY-T/6540-2002 "Lab Testing Method of Drilling and Completion Fluids Damaging Oil Formation", the degree of formation contamination damage and the permeability recovery rate of ultra-deepwater loss-circulation drilling fluid were tested using a dynamic contamination experiment and compared with a loss-circulation drilling fluid prepared using ultra-deepwater drilling fluid + 3% ultrafine calcium carbonate. Different types of cores were selected, and the permeability of cores before and after plugging and plugging removal was measured. The permeability recovery rate was calculated according to the following formula:

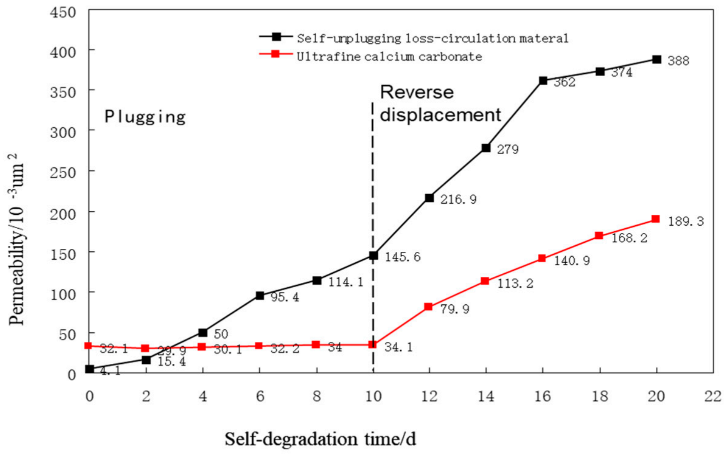
$$\omega = K_1/K_2 \times 100 \quad (3)$$

where ω represents the permeability recovery rate, dimensionless; K_1 represents the permeability after plugging, μm^2; K_2 represents the permeability before plugging, μm^2. In this paper, two types of cores are selected: one is a fractured granite core with a fracture width of 0.2~0.8 mm and the other is a highly permeable sandstone core. The pore distribution was quantitatively tested by using a nanoVoxel-3000 series X-ray three-dimensional CT scanning microscope. The length of the rock sample was 24 mm, the resolution was 17 μm and the scanning mode was wide field scanning.

Figure 7 shows the permeability of different cores before and after plugging. For fractured cores, the permeability was 391.3×10^{-3} μm^2 before plugging, as shown in Figure 7a, and the permeability of the ultra-deepwater plugging drilling fluid with self-degrading loss-circulation material decreased significantly after plugging, and the plugging rate reached 98.9%. After reverse displacement, the permeability recovery rate reached 99.15%. The traditional drilling fluid has no self-degrading capability, and the permeability recovery rate after reverse displacement was only 48.7%. For highly permeable sandstone cores, the permeability was 261.3×10^{-3} μm^2 before plugging, as shown in Figure 7b, and the ultra-deepwater plugging drilling fluid and traditional drilling fluid had similar trends. After plugging with the ultra-deepwater plugging drilling fluid, the plugging rate reached 98.8%, and the permeability recovery rate after reverse displacement was 96.9%. The above results show that ultra-deepwater plugging drilling fluid with the self-degrading loss-circulation material has an excellent self-plugging ability, which can reduce plugging damage to reservoir pores and throats and does not need other treatment measures, such as acidification, biological enzyme, gel breaker, etc., which simplifies operational procedures and reduces cost.

Considering that the plugging performance and permeability recovery of the ultra-deepwater plugging drilling fluid in highly permeable cores are lower than those of fractured cores, the microstructure and pore conditions of sandstone cores before and after plugging and plugging removal were analyzed. Figure 8 shows the SEM scanning results of the microstructure of highly permeable sandstone cores before and after plugging with the ultra-deepwater plugging drilling fluid and after plugging removal. It can be concluded that self-degrading loss-circulation material can form a dense plugging layer in the pore throat, which is beneficial for plugging pores or fractures while drilling. At the same time, the self-degrading loss-circulation material can automatically undergo a degradation reaction at the pore throat of the reservoir. Moreover, the micromolecule organic acid formed by degradation is also helpful in removing plugging in the formation, and it is also very helpful in enhancing the connectivity of the reservoir. However, in small pores, there may be a small amount of residual loss-circulation material, which may be the reason for the slight decrease in the plugging removal rate.

Using an X-ray three-dimensional microscope, the pore distribution of the highly permeable core before plugging and after plugging removal was quantitatively tested, and the pore structure was analyzed. The results are shown in Table 3. In the original rock core, with an increase in the pore equivalent diameter, the number of pores increased at first and then decreased. The pore size was mainly between 60 and 120 μm. After plugging, compared with the original one, the number of medium-diameter pores was greatly reduced, the number of large-diameter pore sizes was obviously reduced and the number of small-diameter pore sizes was greatly increased. The number distribution of pore sizes shows a trend of rapid decline with a gradual increase in the pore equivalent diameter, and the overall pore size is mainly 30–60 µm. This shows that the loss-circulation material has effectively plugged large and medium pores in the rock core, and although there are some micro-pores, it still meets the requirements of plugging while drilling. After plugging removal, it can be seen from pore screening statistics that the number of medium and large pore sizes has obviously increased, which is close to the number of original cores. However, the number of pores with sizes between 30 and 60 µm is obviously less than the original core, and the number of pores with a size greater than 30 µm is slightly more than that of the original core, which may be due to the fact that some loss-circulation material still exists in a small number of pores with sizes between 30 and 60 µm. Based on the pore size contribution, pores of 120–240 µm are the majority, accounting for 45.72% of the total pore volume. By comparing the pore equivalent diameter before plugging and after plugging removal, it can be seen that the self-unplugging loss-circulation material has a good plugging removal effect, and the core permeability recovers after plugging removal.



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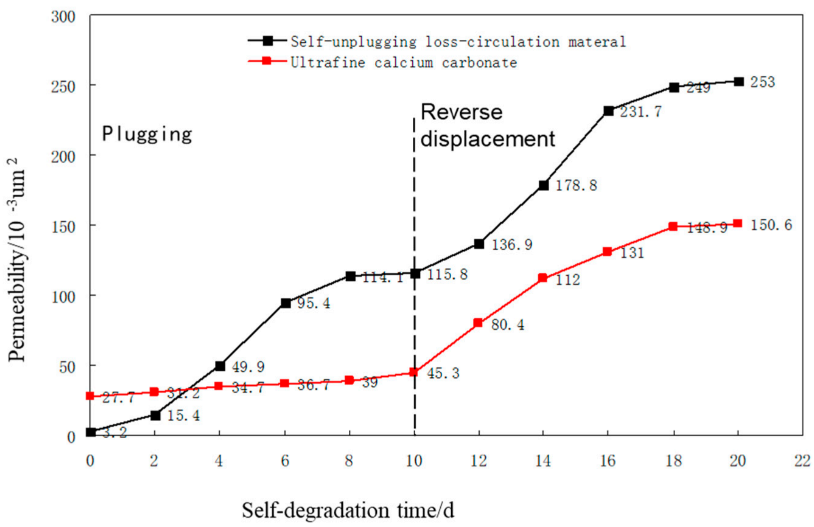

(**b**)

Figure 7. Permeability test of core contamination. (**a**) Fractured granite cores; (**b**) highly permeable sandstone cores.

Figure 8. The microstructure of natural sandstone before plugging and after plugging removal.

Table 3. Percentage of pore equivalent diameter range in total pore volume.

	Pore Equivalent Diameter Range	≤30	30~60	60~120	120~240	240~480	480~960	>960
Before plugging	Number	166,758	278,838	211,153	51,132	4244	99	1
	Percentage of total pore volume	0.40%	5.22%	26.05%	41.64%	23.24%	3.25%	0.20%
After plugging	Number	317,399	216,848	118,207	25,961	1432	9	0
	Percentage of total pore volume	1.41%	7.91%	30.72%	44.09%	15.21%	0.66%	0%
Plugging removal	Number	177,628	236,249	210,698	50,875	3927	99	1
	Percentage of total pore volume	0.6%	4.95%	25.90 %	40.89%	23.27%	4.19%	0.2%

The above research shows that after drilling to total depth, the filter cake layer formed by self-degrading loss-circulation material will degrade over a certain period of time so that the filter cake layer or particles that penetrate into the reservoir can automatically degrade; then, the reservoir can be automatically and efficiently unplugged so that recovery rate of reservoir permeability is high, which significantly improves the productivity of oil and gas wells.

4. Conclusions

(1) A novel self-degrading loss-circulation material was prepared, which can spontaneously degrade into organic weak acid molecules, inorganic small molecules, etc., within a certain time range, and its degradation rate will increase with an increase in temperature and a decrease in material particle size; at 95 °C and pH = 14, the loss-circulation material can completely degrade in 5 days.

(2) The novel self-degrading loss-circulation material has great compressive strength, which can be processed into particles with different particle sizes. When added to the drilling fluid, it can achieve a good plugging effect. With a 3% additive amount, the bearing capacity of 0.2 mm and 1.0 mm micro-fractures can reach 7.6 MPa and 6.9 MPa, respectively, which can prevent drilling fluid from loss-circulation in highly permeable reservoirs and fractured reservoirs, and it has good compatibility with the deepwater drilling fluid.

(3) The novel self-degrading loss-circulation material and filter cake of the ultra-deepwater drilling fluid can degrade automatically after drilling to total depth without other auxiliary treatment measures, such as acidification, biological enzymes, gel breakers, etc. It can basically realize plugging removal. The permeability recovery rates of highly permeable reservoirs and fractured reservoirs reach 99.15% and 96.9% after 10 days, respectively.

(4) The novel self-degrading loss-circulation material has fine compatibility with ultra-deepwater drilling fluid and formation, and its filter cake can automatically degrade, which can significantly improve the effect of reservoir protection. Moreover, this material has no contamination or toxic effect on the environment and can provide technical support for plugging while drilling and reservoir protection for ultra-deepwater granite reservoirs in the South China Sea.

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