Synthesis of a Flexible Particle as a Lost Circulation Material in Drilling Fluids

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Abstract. Lost circulation is always a worldwide problem which is difficult to solve in the process of oil and gas exploration and development. In recent years, lots of lost circulation materials (LCM) and technical aspects of the research work have been made at home and abroad. However, most LCM cannot has good deformability, swelling, temperature resistance, salt resistance and other properties at the same time, resulting in poor lost control and repeated leakage occurred. In order to solve the defects of these LCM, the flexible particle lost circulation material for drilling fluids was developed. The lost circulation material is designed from molecular structure. Through the orthogonal experiment, a flexible particle lost circulation material with a temperature resistance above 150 °C is developed. And the swelling increase of the lost circulation material increased by 7 times in salt water, and it had better water swelling and toughness at high temperature. And at the same time, the water swelling of the lost circulation material has a certain delay. The matching between the particle size of the flexible particle lost circulation material and the sand bed is studied to determine the optimum particle size of lost circulation material.

1. Introduction

Oil and gas resources are an important part of energy, and its development is related to the country's economic and social sustainable development and energy security[1]. Due to the complexity of the formation and the influence of mining methods during oil and gas exploitation, the pressure bearing capacity of some strata has been greatly reduced, and the problem of lost circulation has become an important safety issue[2]. The problem of lost circulation will not only lead to the loss of a large amount of drilling fluids, but also cause great damage to the reservoir[3]. And it will also cause problems such as blowout, instability of the borehole wall, collapse of the borehole wall, and even a blowout accident. Therefore, the control of lost circulation has always been an important link in the oil and gas exploration and development process, and has attracted the attention of oil workers[4].

In oil and gas exploration and development, the most common technical means of controlling leakage is to add a lost circulation material to the drilling fluids[5]. However, the deformability and expansion of the conventional plugging material are poor, which makes it difficult for the lost circulation material to enter the lost formation, resulting in low plugging strength and poor sealing effect. With further research by experts and scholars, flexible particles have excellent deformability and expansion, but the traditional flexible materials have low strength and poor temperature resistance after water swelling, which severely limits their application in plugging materials[6]. In this paper, a new type of flexible
material lost circulation material is developed to make up for the deficiency of traditional flexible particle lost circulation material.

2. Development of flexible particle lost circulation material for drilling fluids
The flexible particle lost circulation material is essentially a water absorbing resin, so it is suitable for the preparation method of the water absorbing resin. There are many preparation methods of water absorbing resin, but there are three main methods: solution polymerization, reverse suspension polymerization and reverse emulsion polymerization[7].

The main raw materials of this polymerization experiment, such as AM, AA and AMPS, are water-soluble. Moreover, when the polymerization reaction is carried out with water as the solvent, the process is simple, the reaction heat dissipation is fast, the reaction temperature is easy to control, which is conducive to avoiding local high temperature, and the obtained molecular weight of the product is more uniform. In addition, aqueous solution polymerization is the most widely used and mature polymerization process, so this paper chooses aqueous solution polymerization[8].

2.1. Monomer selection
According to the analysis of molecular structure design and a large number of literature research, three main monomers, acrylamide (AM), acrylic acid (AA) and 2-acrylamide-2-methylpropionic sulfonic acid (AMPS), are initially selected. And four synthetic schemes were designed. By testing the water swell ability, toughness, etc. of the product, the possibility of a synthetic scheme is explored, thereby determining the synthesis monomer of the soft particle lost circulation material, and then performing a subsequent orthogonal test. Four of the synthetic schemes are as follows:

1#: AMPS:AM=0:20; 2#: AMPS:AM=2:20; 3#: AMPS:AM=4:16; 4#: AMPS:AA=2:1:17

2.2.1. Preliminary synthesis experiment. During the synthesis experiment, it was found that the concentration of CaCl₂ solution had a great influence on the product properties. Therefore, a synthetic experiment with CaCl₂ concentrations of 3%, 4%, 5%, and 6% was performed. The water swelling property and toughness after aging of the product are preferred indicators, and the synthesis scheme is evaluated, and a preferred scheme is preferred for subsequent experiments. The aging conditions used in the experiment were 150 °C, and the hot rolling was performed for 16 h.

Obtained through experimental that as the concentration of CaCl₂ increases, the water absorption expansion ratio of the products of each scheme decreases after aging, and when the concentration of CaCl₂ is 3%, only the products of 1# and 4# are dissolved after aging. This indicates that the addition of AMPS does improve the high temperature resistance of the product to some extent. As the concentration of CaCl₂ increases, the toughness of the product increases and the change of 1# is more obvious. It can be concluded that increasing the concentration of CaCl₂ can improve the high temperature resistance and toughness of the product. At the same time, under the 6% CaCl₂ concentration, the 1# product swelled by 49.25 times and the toughness was D, which was better than other schemes. Moreover, the 1# solution is more cost effective than other solutions, and there are fewer experimental steps. In order to verify the possibility of 1#, it was found that at 12% CaCl₂ concentration, the toughness of the product after aging was E, which satisfied the synthesis requirements. Therefore, 1# was selected as the primary selection scheme for subsequent experiments.

2.2.2. Orthogonal experimental analysis. The monomer concentration, the amount of functional monomer X, the concentration of CaCl₂, the crosslinking agent, the initiator, the amount of bentonite, the reaction temperature, etc. all have an effect on the performance of the flexible particle lost circulation material. Therefore, this experiment designed 18 sets of experiments according to L₁₈ (3⁷). Seven factors including the monomer concentration, the addition amount of functional monomer X, the concentration of CaCl₂, the crosslinking agent, the initiator, the amount of bentonite, and the reaction temperature were taken as factors to be investigated, and three factors were taken as factors to investigate. Three levels of each factor were selected for orthogonal experiments. In this experiment, the water swollability
and toughness of the product after aging are selected as the preferred indicators. The orthogonal experimental table is as follows:

| Table 2-1 L₁₈ (3⁷) factor-level |
|---------------------------------|
| Factor level | A | B | C | D | E | F | G |          |          |          |
|---------------|---|---|---|---|---|---|---|----------|----------|----------|
|   | Functional monomer X (%) | Monomer concentration (%) | Crosslinking agent (ml) | Initiator (ml) | Bentonite (%) | Concentration of CaCl₂ (%) | Reaction temperature (℃) |
| 1  | 2 | 15 | 2 | 1.4 | 14 | 7 | 40 |
| 2  | 3 | 20 | 3 | 1.6 | 18 | 8 | 50 |
| 3  | 4 | 25 | 4 | 1.8 | 22 | 9 | 60 |

Note 1: Functional monomer X is added as a percentage of functional monomer X to the total mass of the monomer; the crosslinker concentration is 0.01 g/ml; the initiator concentration is 0.02 g/ml, and the CaCl₂ solution used is 100 ml.

| Table 2-2 L₁₈ (3⁷) experimental results |
|-----------------------------------------|
| Experiment number | A | B | C | D | E | F | G | Water absorption coefficient (g/g) | Stretch multiple |
|-------------------|---|---|---|---|---|---|---|-----------------------------------|-----------------|
| 1                 | 1 | 1 | 1 | 1 | 1 | 1 | 1 | 6.25                             | Easy to break   |
| 2                 | 1 | 2 | 2 | 2 | 2 | 2 | 2 | 17.95                            |                 |
| 3                 | 1 | 3 | 3 | 3 | 3 | 3 | 3 | 16.70                            |                 |
| 4                 | 2 | 1 | 1 | 2 | 2 | 3 | 3 | 4.33                             | Easy to break   |
| 5                 | 2 | 1 | 1 | 3 | 3 | 1 | 1 | 13.47                            |                 |
| 6                 | 2 | 3 | 3 | 1 | 1 | 2 | 2 | 49.93                            |                 |
| 7                 | 3 | 1 | 2 | 1 | 3 | 2 | 3 | 5.18                             | Easy to break   |
| 8                 | 3 | 2 | 3 | 2 | 1 | 3 | 1 | 8.02                             | 3.5             |
| 9                 | 3 | 3 | 1 | 3 | 2 | 1 | 2 | 41.67                            | 2.7             |
| 10                | 1 | 1 | 3 | 3 | 2 | 2 | 1 | 4.03                             |                 |
| 11                | 1 | 2 | 1 | 1 | 3 | 3 | 2 | 7.62                             | 3.2             |
| 12                | 1 | 3 | 2 | 2 | 1 | 1 | 3 | 70.25                            | Easy to break   |
| 13                | 2 | 1 | 2 | 3 | 1 | 3 | 2 | 1.99                             | 5.2             |
| 14                | 2 | 2 | 3 | 1 | 2 | 1 | 3 | 15.83                            | 5.3             |
| 15                | 2 | 3 | 1 | 2 | 3 | 2 | 1 | 17.00                            | Easy to break   |
| 16                | 3 | 1 | 3 | 2 | 3 | 1 | 2 | 5.12                             | 2.8             |
| 17                | 3 | 2 | 1 | 3 | 1 | 2 | 3 | 19.55                            |                 |
| 18                | 3 | 3 | 2 | 1 | 2 | 3 | 1 | 8.80                             | Easy to break   |

| Table 2-3 L₁₈ (3⁷) experimental results |
|-----------------------------------------|
| A | B | C | D | E | F | G |
|---|---|---|---|---|---|---|
| Functional monomer X (%) | Monomer concentration (%) | Crosslinking agent (ml) | Initiator (ml) | Bentonite (%) | CaCl₂ (%) | Temperature (℃) |
|-----------------|-----------------|-----------------|-----------------|-----------------|----------|-----------------|
| K1 | Water swelling ratio | 20.47 | 4.48 | 16.07 | 15.60 | 26.00 | 25.43 | 9.60 |
|    | Tensile ratio        | 1.9   | 3.6  | 2.2   | 2.4   | 2.3   | 2.0   | 2.2 |
| K2 | Water swelling ratio | 17.09 | 13.74| 19.61 | 20.45 | 15.44 | 18.9  | 20.71|
|    | Tensile ratio        | 2.4   | 1.7  | 2.4   | 2.1   | 2.5   | 1.8   | 2.4 |
| K3 | Water swelling ratio | 14.73 | 34.06| 16.61 | 16.24 | 10.85 | 7.91  | 21.97|
|    | Tensile ratio        | 2.4   | 1.4  | 2.2   | 2.2   | 1.9   | 3.0   | 2.1 |
| R  | Water swelling ratio | 5.74  | 29.57| 3.54  | 4.84  | 15.15 | 17.52 | 12.38|
|    | Tensile ratio        | 0.5   | 2.1  | 0.2   | 0.3   | 0.6   | 1.2   | 0.3 |

Note 2: K1, K2, and K3 represent the average of the performance indicators corresponding to the number of levels 1, 2, and 3, respectively; R represents the range.

By comparing the R value of each factor affecting the water swell ratio and the tensile ratio after the aging of the flexible particle lost circulation material, the influence of each factor on the product performance can be obtained. In the case of comprehensive consideration of the performance of the lost circulation material, it is considered that the toughness of the lost circulation material is more important, so the water swell expansion ratio of the product is considered again on the premise that the stretching factor is prioritized. Therefore, the experimental conditions for the polymerization were selected as follows: monomer concentration 15%, CaCl2 concentration 9%, bentonite addition 18%, functional monomer X addition 3%, crosslinker addition 2ml, initiator addition 1.6ml, reaction temperature 60 C.

3. Evaluation of plugging performance of flexible particle lost circulation material

3.1. Salt resistance evaluation

The toughness and water swellability of the flexible particle lost circulation material under different absorption media were measured. The toughness of the flexible particle lost circulation material was E, which was basically unchanged, as measured by different NaCl and CaCl2 concentrations. Figure 3-1 and Figure 3-2 show the effect of different NaCl and CaCl2 concentrations on the water swellability of flexible particle lost circulation materials at room temperature.
It can be seen from Figure 3-1 and Figure 3-2 that as the salt concentration increases, the water swell expansion ratio of the flexible particle lost circulation material increases. And the swelling increasement of the lost circulation material increased by 7 times in salt water, it can meet the requirements of leakage prevention and plugging operation under high salinity.

3.2. Sand bed experiment
Due to the anisotropy of the formation, in order to get closer to the actual situation of the well, it is necessary to select different sand layers for the plugging experiment, determine the plugging effect, and find out the sealing law of the flexible particle lost circulation material.

In this paper, the FA.BX sand bed plugging instrument is used to carry out the sand bed experiment. In order to find out the sealing law of the flexible particle lost circulation material on the sand bed, this paper selects the lost circulation material with different particle size as the sand bed experiment. Thereby, the plugging effect of the flexible particle lost circulation material and the optimal plugging particle size required for the sand bed of each particle size are determined.

The base mud + 1% flexible particle lost circulation material was used as the sand bed experiment. And the experimental results are shown in Figure 3-3 and Figure 3-4:

![Figure 3-1: The effect of the concentration of NaCl on water-swelling property of product](image1)

![Figure 3-2: The effect of the concentration of CaCl₂ on water-swelling property of product](image2)

![Figure 3-3: Penetration depth of flexible particles with various particle size distributions on 40-60 mesh sand bed](image3)

![Figure 3-4: Penetration depth of flexible particles with various particle size distributions on 20-40 mesh sand bed](image4)
It can be seen from Figure 3-3 that the 100-200 mesh flexible particle lost circulation material is better for the 40-60 mesh sand bed and 80-100 mesh. And the Figure 3-4 shows that the lost circulation material with a better sealing effect on the 20-40 mesh sand bed has a particle size of 80-100 mesh. Moreover, when the flexible lost circulation material particles are between 100-200 mesh, the drilling fluids will completely pass through the sand bed.

When testing the sealing effect of the flexible particle lost circulation material on the 10-20 mesh sand bed, it was found that the base mud + 1% flexible particle lost circulation material had no blocking effect on the 10-20 mesh sand bed. Therefore, consider the different particle size of the plugging, do the sand bed experiment, the experimental results are shown in Table 3-1.

| Flexible particle lost circulation material with different particle size grades | Sand bed penetration depth /cm |
|---|---|
| 1% (40-60 mesh) +1% (60-80 mesh) | 9.6 |
| 1% (40-60 mesh) +1% (80-100 mesh) | 9.8 |
| 1% (40-60 mesh) +0.5% (60-80 mesh) +0.5% (80-100 mesh) | 6.8 |
| 0.8% (40-60 mesh) +0.6% (60-80 mesh) +0.6% (80-100 mesh) | 7.7 |
| 1.2% (40-60 mesh) +0.4% (60-80 mesh) +0.4% (80-100 mesh) | 9.2 |
| 1.6% (40-60 mesh) +0.2% (60-80 mesh) +0.2% (80-100 mesh) | 10.2 |
| 1% (40-60 mesh) +0.6% (60-80 mesh) +0.4% (80-100 mesh) | 7.2 |
| 1% (40-60 mesh) +0.4% (60-80 mesh) +0.6% (80-100 mesh) | 7.5 |
| 1% (40-60 mesh) +0.5% (60-80 mesh) +0.4% (80-100 mesh) +0.1% (100-200 mesh) | 6.4 |

As seen from Table 3-1, when the amount of flexible particle lost circulation material is 2%, 1% (40-60 mesh) + 0.5% (60-80 mesh) + 0.4% (80-100 mesh) + 0.1% (100-200 mesh) has a depth of 6.4 cm, which is the best for the 10-20 mesh sand bed.

4. Conclusions
Through orthogonal experiment analysis, the optimal synthetic formula of flexible particle lost circulation material was determined as follows: in 100 ml CaCl₂ solution, monomer concentration 15%, CaCl₂ concentration 9%, bentonite addition 18%, functional monomer X addition 3%, crosslinker addition 2 ml, initiator addition 1.6 ml, reaction temperature 60°C. The matching of flexible particle lost circulation material with different sand beds was studied by sand bed plugging experiment. The particle size of the lost circulation material was determined as: 1% (40-60 mesh) + 0.5% (60-80 mesh) + 0.4% (80-100 mesh) + 0.1% (100-200 mesh).

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