The performance of a novel inorganic organic composite drilling fluids loss reducer AM/AA/GO

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Abstract. In this paper, an inorganic/organic water-based drilling fluids reducer AM/AA/GO was prepared by using acrylamide (AM), acrylic acid (AA) and graphene oxide (GO) as raw material. Performance evaluation showed that the AM/AA/GO has an excellent filtration loss reduce performance. It has a good salt resistance performance and its temperatures resistance ability was up to 140 °C. The comprehensive results showed that AM/AA/GO is expected to be used in anti-collapse water-based drilling fluids.

Introduction

In the shale reservoir, the hydration expansion dispersion of wellbore is easily lead to the borehole collapse, block falling, sticking and other drilling projects in drilling engineering. Therefore, high requirements are put forward drilling fluids for loss reduce, shale inhibition and plugging ability [1]. The drilling fluids cake is the first drilling barrier to prevent the intrusion of water into oil and gas formation, therefore, the quality and the effective degree of drilling fluids cake will be directly affect the stability of wellbore, which is one of the most concerned indexe of drilling fluid performance to the drilling researcher [2]. As a fluids loss reducing agent, which participates in the cake formation and as an important part of mud cake, water-based drilling fluids loss reducer can greatly affects the filtration volume loss of drilling fluids [3, 4].

For a long time, there are many fluids loss reducers products, most of which are binary, ternary, quaternary, and even ternary co-polymers. The monomers are mostly acrylic acid (AA), acrylate (MA), acrylamide (AM) and functional monomer containing high-temperature-resistant function groups. These co-polymers undertake the task of reducing filtration loss within a certain period time and have promoted the progress of drilling engineering [5-7].

Nanomaterial have their own unique surface characteristics and thermal conductivity, and they have unparalleled material advantages in drilling fluid for rheology control, thermal conductivity and filtration loss performance, and have shown good development prospects in drilling fluids. Integrating the respective advantages of polymers and nanomaterial, nanoparticle/polymer composites have been initially explored in drilling fluids and have shown excellent performance [8]. But at the same time, it
is an indisputable fact that nanomaterial are easy to re-unite in the aqueous solution and are sensitive to salt, which has led to the limitation of their large-scale applications [9, 10].

Based on the above research background, in this paper, an novel water-based drilling fluids reducer AM/AA/GO was prepared by using acrylamide (AM), acrylic acid (AA) and graphene oxide (GO) as raw material. The performance of AM/AA/GO was evaluated by using filtration loss experiments.

1. Experimental

1.1. Materials

AM，AA，NaOH，(NH4)2S2O8，NaHSO3，Aladdin reagent，all the above reagents are analytically pure; graphite powder, Qingdao Chongyang Graphite Co., Ltd; Bentonite was purchased from Xinjiang Xiazi Street Bentonite Co., Ltd.; shale samples were taken from Chongqing, Drilling fluids materials were all purchased from Shandong Deshunyuan Co., Ltd.

1.2. Preparation of AM/AA/GO

1.7 g acrylic acid and 30 mL deionized water added into a round-bottomed flask, the solutions was adjusted to 7 by 25% NaOH, and 3.7 g acrylamide was add by following. Hereafter, the initiator K2S2O8 and NaHSO3 (n: n = 1: 1), which the initiator concentration is 0.8 wt% relative to the total monomer amount were added. The polymerization was carried out at 50 °C for 0.5 h while stirring, and then isopropanol was added to the solution, polymerization was preceded at 70 °C. The resulting product was obtained by repeatedly washing with ethanol to remove monomers, and the polymer AM/AA was obtained. Add 1g AM/AA and 20 mL deionized water in a round-bottomed flask, and then raised the temperature to 30 ° C, slowly add 0.7 g OP-10, and then add 0.75 g GO to the round-bottomed flask. Ultrasonic reaction was carried out for 2 h at a sealed condition and AM/AA/GO was obtained after washing, drying and shearing granulation.

1.3. Drilling fluids performance evaluation

1.3.1. Based muds preparation.

Add 10 L distilled water into the iron container and 400 g calcium bentonite was added in the mixing state. After that, 2.4g Na2CO3 was added and maintained the based muds 48 h.

1.3.2. Drilling fluids performance tests.

The performance tests of drilling fluids was according to the standard SY / T 5621-1993 of China.

2. Results and discussion

2.1. The effect of AM/AA/GO on rheological properties of based muds

Different amount of AM/AA/GO was added to the based muds which has been pre-hydrated for 48 h, and the effect result of AM/AA/GO on the rheology of based muds are shown in Table 1.

| system                  | AV/mPa·s | PV/mPa·s | YP/Pa  | YP/PV  | Gel/Pa/Pa |
|-------------------------|----------|----------|--------|--------|-----------|
| Based muds              | 5.5      | 5        | 0.5    | 0.1    | 1/1       |
| Based muds +0.1% AM/AA/GO | 7        | 5        | 2      | 0.4    | 1/1       |
| Based muds +0.2% AM/AA/GO | 10       | 7        | 3      | 0.4    | 1/2       |
| Based muds +0.5% AM/AA/GO | 12       | 8        | 4      | 0.5    | 2/3       |
| Based muds +0.8% AM/AA/GO | 15       | 10       | 5      | 0.5    | 5/8       |
| Based muds +1.0% AM/AA/GO | 23       | 14       | 9      | 0.6    | 7/13      |
It can be seen from Table 1 that with the increase addition amount of AM/AA/GO, the viscosity of the based muds system show a gradual rise trend. The rise of shear force is very conducive to the suspension of drilling fluid to drilling cuttings, which can bring cuttings out of the wellbore. When the dosage was more than 0.8%, the viscosity of based muds has been improved obviously. It can be seen that the most suitable dosage of AM/AA/GO in the based muds was 0.8%.

2.2. Effect of salt on rheological property of AM/AA/GO based muds

When drilling to the salt gypsum layer, the performance of drilling fluids will be changed due to the invasion of salts. In addition, AM/AA/GO contains graphene oxide which is sensitive to salts, so it is necessary to investigate its salt resistance ability. Figure 1 and Figure 2 show the effect of NaCl and CaCl₂ on the rheological property of 0.8% AM/AA/GO based muds, respectively.

![Figure 1. Effect of NaCl on rheological property of AM/AA/GO based muds](image)

![Figure 2. Effect of CaCl₂ on rheological property of AM/AA/GO based muds](image)

Inorganic salts can ionize cations in water, and cations have a great influence on the branched chain extension of polymers. The amide group of AM/AA/GO can hydrolyze carboxylic acid anion in aqueous solution. In addition, AM/AA/GO itself has carboxylic acid group. The salt in drilling fluid can make the double electric layer and hydration layer of carboxylic acid thin, weaken the repulsion of carboxylic acid group, and make the molecular chain of AM/AA/GO curl, thus losing the rheological property. It can be seen from Figure 1 and Figure 2 that with the addition increase of salts, the viscosity of AM/AA/GO based muds showed a trend of attenuation, but CaCl₂ has a greater impact on it. When the dosage of NaCl is 12%, the apparent viscosity of AM/AA/GO based muds has been reduced to 2 mPa·s, when the dosage of CaCl₂ is 1.0%, the suspension performance of the system has been lost.

2.3. Effect of salt on rheological property of AM/AA/GO based muds

On the one hand, temperature will affect the space expansion of polymer, on the other hand, it will promote the decomposition of polymer at high temperature. The effect of temperature on the rheological properties of 0.8% AM/AA/GO based muds was investigated in the laboratory, as shown in Table 2.

| Condition     | AV/mPa·s | PV/mPa·s | YP/Pa | YP/PV | Gel/Pa/Pa |
|---------------|----------|----------|-------|-------|-----------|
| Room temp     | 15       | 10       | 5     | 0.5   | 5/8       |
| 100°C×16h     | 19       | 12       | 7     | 0.6   | 6/8       |
| 120°C×16h     | 15       | 9        | 6     | 0.7   | 4/5       |
| 130°C×16h     | 13       | 8        | 5     | 0.6   | 3/4       |
| 140°C×16h     | 9        | 6        | 3     | 0.5   | 1/2       |
| 150°C×16h     | 5        | 3        | 2     | 0.7   | 1/1       |
It can be seen from Table 2 that the rheological properties of 0.8% AM/AA/GO based muds show a downward trend with the increase of hot rolling temperature. However, the rheological property of drilling fluid increased slightly after 100 °C × 16h. The reason may be that under 100 °C, the polymer chain of AM/AA/GO has not been fully extended, and AM/AA/GO has not fully developed its own viscosity increasing effect. With the further increase of hot rolling temperature, AM/AA/GO has begun to decompose, the spatial network state between polymers has been destroyed to a certain extent, and the viscosity of AM/AA/GO has begun to decline. When the hot rolling temperature reached to 140 °C, the rheological property of the system has been completely destroyed. It can be seen that the temperature resistance of AM/AA/GO in the based muds system can be as high as 140 °C.

2.4. Effect of salt on rheological property of AM/AA/GO based muds

The filtration reduction performance of AM/AA/GO was studied in 4% based muds, and the result is shown in Figure 3.

![Figure 3. Fluids loss performance study of AM/AA/GO](image)

**Figure 3.** Fluids loss performance study of AM/AA/GO

AM/AA/GO was combination of inorganic and organic materials, its physical properties show the dual characteristics of the rigidity of inorganic particles and the deformability of polymer. It can reduce the porosity of mud cake by using the role of plugging and bridging of inorganic particles, and then play a role of filtration reduction. It can be seen from Fig. 3 that the filtration capacity of based muds was 15.3 mL. With the increase of the addition of filtrate reducer AM/AA/GO, the filtration amount of polymer based muds has been significantly reduced, when added mount of AM/AA/GO was 0.8%, the water loss was only 5.4 mL. AM/AA/GO / % FL / mL

3. Conclusion

In this paper, a novel water-based drilling fluids reducer AM/AA/GO was prepared with AM and AA and GO. The performance evaluation results showed that AM/AA/GO has a nice water loss reduction performance. AM/AA/GO can rely on inorganic / organic components to block the muds cake, reduced the water loss of drilling fluids and maintain the stability of the wellbore.

Acknowledgments

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