Synthesis of a Novel Water-Based Drilling Fluids Loss Reducer

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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. Structural of AM/AA/GO was characterized via FT-IR, GPC and TGA-DSC. GPC results showed that the molecular weight of AM/AA/GO was 23503 g/mol and the TGA-DSC confirmed the excellent temperature resistance ability of AM/AA/GO.

Keywords. Shale gas, water based drilling fluids, loss reducer.

1. Introduction
In the shale reservoir, the hydration expansion dispersion of well is easily lead to the borehole collapse, block falling, sticking and other drilling projects in drilling engineering. Therefore, high requirements are put forward for drilling fluids loss, shale inhibition and plugging ability of drilling fluids [1-3]. 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 indexes of drilling fluid performance to the drilling researcher [4, 5]. 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 amount of filtration volume [6].

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

Nanomaterial have their own unique surface characteristics and thermal conductivity, and they have unparalleled material advantages in drilling fluid 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. 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 [8-10].
Based on the above research background, in this paper, a 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. Structural of AM/AA/GO was characterized via FT-IR, GPC and TGA-DSC.

2. Experimental

2.1. Materials
AM, AA, NaOH, (NH₄)₂S₂O₈, NaHSO₃, NaNO₃, KCl, K₂MnO₄, BaCl₂, Aladdin reagent, all the above reagents are analytically pure; 98% H₂SO₄, 35% HCl, Shanghai Aladdin Biochemical Technology Co. Ltd. graphite powder, Qingdao Chenyang Graphite Co., Ltd.

2.2. 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.

2.3. Structure Characterization
FT-IR spectra were recorded via a WQF-520 Fourier transform infrared spectrometer in the wave number range of 4000-5000 cm⁻¹. GPC was utilized to measure the molecular weight by using an Alliance e2695 instrument (Waters, USA). The injection volume and operation hours were 50 μL and 90 min, respectively. TGA-DSC tests were acquired on a simultaneous TGA-DSC (METTLER TOLEDO, Swiss) instrument under nitrogen atmosphere flow (40 mL min⁻¹) with the heating rate of 10 °C min⁻¹.

3. Results and Discussion

3.1. FT-IR
The FT-IR characterization of AM/AA/GO was performed by using a tableting method, the measurement wavelength was range from 4000-500 cm⁻¹, and the result is showed in figure 1.

![Figure 1. The infrared spectra of AM/AA/GO.](image-url)
On the AM/AA/GO infrared spectrum curve as can be seen in figure 1, we have observed that a broad absorption peak at 3300 cm\(^{-1}\), which is the bending vibration absorption peak of NH\(_2\), and the narrow absorption peak at 1720 cm\(^{-1}\) is the stretching vibration absorption peak of NH\(_2\). 1460 cm\(^{-1}\) and 1263 at cm\(^{-1}\) is the absorption peak of -COOH on the side chain of AM/AA/GO. FT-IR spectrum result showed that the monomers has participated in the copolymerization, and physicochemical adsorption was occurred between the polymer and GO.

3.2. TGA-DSC
Decomposition of drilling fluids treatment agents in deep wells will have a very adverse effect on drilling fluids performance. As the temperature increases, it will inevitably decompose as AM/AA/GO is composed of polymers and GO. The TGA-DSC result was shown in figure 2.

![Figure 2. TGA-DSC of AM/AA/GO.](image)

It can be seen from figure 2 that the weight loss curve of AM/AA/GO can be roughly divided into three stages between 40 °C and 800 °C. The weight loss rate of 40 °C to 100 °C in the first stage is 3.67%, which corresponds to desorption of adsorbed water in the polymer. The mass loss in this stage is mainly the weight loss of the side chain functional groups in the polymer. The mass loss in the 400 °C to 800 °C heating stage in the last stage is due to the decomposition of AM/AA/GO. It can be found from the experiments that AM/AA/GO is expected to be used in high temperature drilling.

3.3. GPC
It is well known that the molecular weight of polymers has a large effect on the rheological property of drilling fluids. The molecular weight distribution of AM/AA/GO was analyzed by using a smart gel permeation chromatography.

From the molecular weight distribution test result in figure 3, we can see that AM/AA/GO has a narrow molecular weight distribution, with an average molecular weight of 23503 g/mol. Its molecular weight is moderate, which can block mud cake and reduce filtration loss, and has little effect on other properties of drilling fluids.

4. Conclusion
In this paper, a novel water-based drilling fluids reducer AM/AA/GO was prepared with AM and AA and GO. FT-IR, GPC and TGA-DSC characterization confirmed that the target product had been formed, its molecular weight was moderate and its temperature resistance was nice. The results showed that PAAM can increase the viscosity of aqueous solution and reduce the water loss of drilling fluids and maintain the wellbore stability.
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Figure 3. Measurement of molecular weight distribution of AM/AA/GO.

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References
[1] Gou S, Yin T, Xia Q and Guo Q 2015 RSC Adv. 41 32064
[2] Huang Y, Zhang D and Zheng W 2019 J Appl Polym Sci. 30 47813
[3] Salami O and Plank J 2012 J Appl Polym Sci. 4 1449
[4] Liu X, Liu K, Gou S, Liang L, Luo C and Guo Q 2014 Ind Eng Chem Res. 8 2903
[5] Rafati R, Smith S, Haddad A, Novara R and Hamidi H 2018 J Petrol Sci Eng.161 61-76
[6] Hassani S, Amrollahi A, Rashidi A, Soleymani M and Rayatdoost, S 2016 J Petrol Sci Eng. 146 183-190
[7] Gou S, Yin T, Liu K and Guo Q 2015 New J Chem. 3 2155
[8] Aftab A, Ismail A, Ibupoto Z 2016 Egy J Petrol. 2 291
[9] Aftab A, Ismail A, Ibupoto Z, Akeiber H and Malghani M 2017 Renew Sust Energ Rev. 76 1301
[10] Abdo J and Haneef M 2013 Appl Clay Sci. 86 76