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Study on Ba Element Solidified by Alkaline Heat Treatment in Oil-based Cuttings

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ABSTRACT

Oil-based cuttings from a shale gas well were studied. The effect of reaction temperature and holding time on curing effect of Ba element in oil-based cuttings by heat treatment method. The experimental results show that the addition of NaOH can effectively solidify Ba element in oil-based cuttings during heat treatment. At the same time, with the increase of reaction temperature, the increase of the proportion of NaOH and the increase of holding time, the leaching amount of Ba was lower and lower.

KEYWORDS

Oil-based cuttings; Ba; Alkaline heat treatment; Reaction temperature; Holding time; NaOH

1. INTRODUCTION

Energy has an important strategic position in the national economy. Shale gas, as one of the important clean energy sources, has abundant reserves in China [1]. On September 14, 2016, the National Energy Administration issued the Shale Gas Development Plan (2016-2020) [2] to accelerate shale gas exploration and development and increase clean energy supply. According to China Mineral Resources Report 2020 [3] released by the Ministry of Natural Resources of China, the newly proved geological reserves of Changning, Weiyuan and Taiyang shale gas fields in Sichuan Basin all exceed 1011m3. Due to the reasons of mining technology, a large number of oil-based cuttings will be produced in the mining process, and oil-based cuttings contain a large amount of oil, heavy metals and other harmful substances, which will cause serious pollution to the environment, and have been listed in the National Hazardous Waste List (HW08 category), and all places are required to prohibit the discharge of oil-based cuttings [4-6]. With the continuous growth of China's shale gas industry, the production of oil-based cuttings is also increasing, which has become one of the main problems restricting the development of shale gas in China [7-16]. Therefore, how to prevent the harm of oil-based cuttings, protect the ecological environment, improve the disposal capacity of oil-based cuttings, and realize the "harmless, resource-based and reductive" treatment of oil-based cuttings is the focus of domestic and foreign scholars.

In order to reduce the harm of Ba, the Ba in oil-based cuttings was cured by experimental research method and alkaline heat treatment method. The influence of heat treatment temperature and treatment time on curing was studied, and the feasibility of curing Ba in oil-based cuttings by heat treatment was analyzed.

2. EXPERIMENTAL MATERIALS AND METHODS

2.1. Experimental Materials and Instruments

The main experimental materials are: deionized water, additives, oil-based cuttings.

Main experimental instruments: box-type electric furnace, blast drying oven, horizontal oscillator, X-ray diffraction analyzer, etc.

2.2. Experimental Methods

The curing and leaching characteristics of Ba were obtained by orthogonal experiment by changing reaction temperature, holding time and proportion of NaOH. The experimental scheme is shown in Table 1:

Table 1. Experimental scheme of heat treatment

Influencing factor	Unit		
Temperature	℃	500	600
Holding time	0.5h~4h		

After basic heat treatment, the sample material composition was obtained by X-ray diffractometer (XRD). According to HJ557-2010 "Solid Waste leaching toxicity leaching method horizontal oscillation method", the amount of Ba compound leaching was detected by inductively coupled plasma emission spectrometer (ICP-OES) to determine the curing effect of heat treatment on Ba element in oil-based cuttings.

2.3. Basic Characteristics of Oil-based Cuttings

Figure 1 shows the basic morphology of the experimental sample of oil-based cuttings. As shown in Figure 1, oil-based cuttings are mainly grays and black viscous solids, which are mainly composed of oil-based drilling fluids, underground rock debris and soil, etc. All oil-based cuttings samples in this paper are from a shale gas well in Rongxian County, Zigong, and the oil content of oil-based cuttings samples is about 6% and the water content is about 5%.

Figure 1. Initial morphology of oil-based cuttings

ICP-OES was used to detect all elements of oil-based cuttings, and according to the identification standard of hazardous waste, it was found that oil-based cuttings contained a large amount of Ba, and the leaching amount of Ba was 26.5 times of the standard limit value, which caused serious pollution to the environment after precipitation.

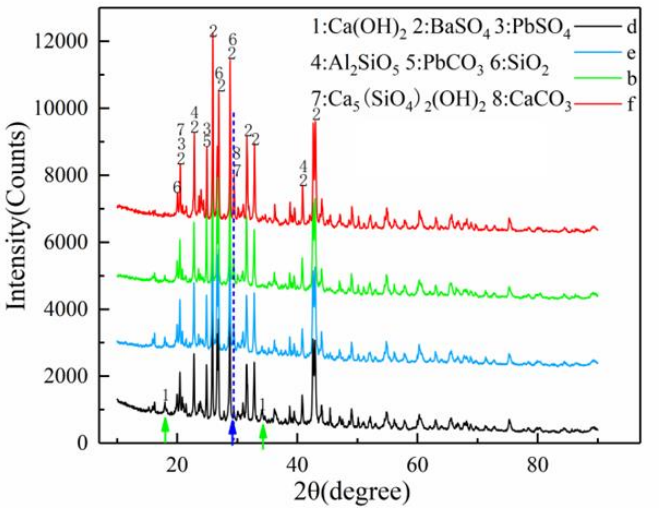
3. ANALYSIS OF EXPERIMENTAL RESULTS

3.1. Influence of Reaction Temperature and Holding Time on Ba Curing

According to the results of the previous part of the study, alkaline heat treatment has a good curing effect of Ba when the proportion of NaOH is 5%. Therefore, in order to analyze the curing effect of reaction temperature and holding time on Ba in alkaline heat treatment oil-based cuttings, the following analysis takes the proportion of NaOH as 5%.

Figure 2 shows the phase changes of samples treated at different temperatures. It can be seen from Figure 2 that the influence of temperature on the phase composition of the sample is lower than that of the additive. In the process of temperature rise, $Ca(OH)_2$ gradually disappears, while $Ca_5(SiO_4)_2(OH)_2$ and $CaCO_3$ diffraction peaks appear.

It can be found from the XRD pattern that Na_2CO_3 and other NA-related compounds are not detected. This phenomenon is mainly because NaOH is in a molten state during the heating process, and the compounds generated by the reaction with CO_2, SiO_2, Al_2O_3 and other oxides are amorphous compounds rather than complete crystal network structures. These amorphous substances can help to further improve the curing effect of heavy metals, thereby reducing their leaching activity [29].



(d: 300℃5%NaOH; e: 400℃5%NaOH; b: 500℃5%NaOH; f: 600℃5%NaOH)

Figure 2. Phase changes at different temperatures (0.5 h)

Figure 3 shows the distribution of Ba on the surface of the sample at different temperatures. Although rising temperature has little effect on the phase composition, after rising temperature, a Ba aggregation phase appears on the surface of the sample, and the change trend is the same as that of adding NaOH, indicating that under the same other conditions, rising temperature can improve the reaction efficiency and make Ba aggregation faster. These clumping phases are where NaOH, in its liquid state, attracts Ba ions.

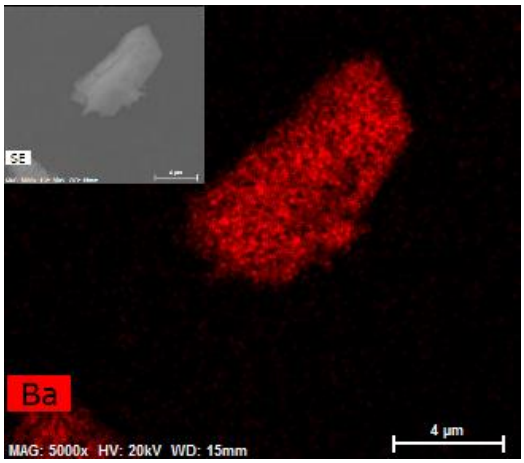


Figure 3. Distribution of Ba on the surface of the sample (treatment duration 0.5h)

Figure 4 shows the change of leaching morphology. It can be found from the figure that more and more white precipitates were observed on the surface of the sample during the temperature rise to 500℃. These white precipitates were the cured products of Ba, indicating that the curing effect of Ba could be improved by increasing temperature, but the change was not obvious when compared with 500℃ and 600℃.

(a) (b)

(a:500℃;b:600℃)

Figure 4. Leaching morphology changes of samples under different temperature conditions

Figure 5 shows the effects of reaction temperature and holding time on the leaching rate of Ba. It can be found that under the conditions of 500℃ the leaching amount of Ba is higher than the initial leaching amount of oil-based cuttings, but with the increase of temperature, the leaching amount of Ba decreases rapidly. When the temperature rises to 600℃, the leaching rate of Ba meets GB5085.3-2007. As the temperature continues to rise, the leaching amount basically does not change.

In terms of the absolute change of the leaching amount, the holding time has relatively little influence on it. After holding for 0.5h, the sample leaching rate already meets GB5085.3-2007, which is 40.27mg/L; if holding for 4h, the leaching amount is 14.00mg/L, and the time is extended by 7 times, the leaching rate is only reduced by 65.23%.

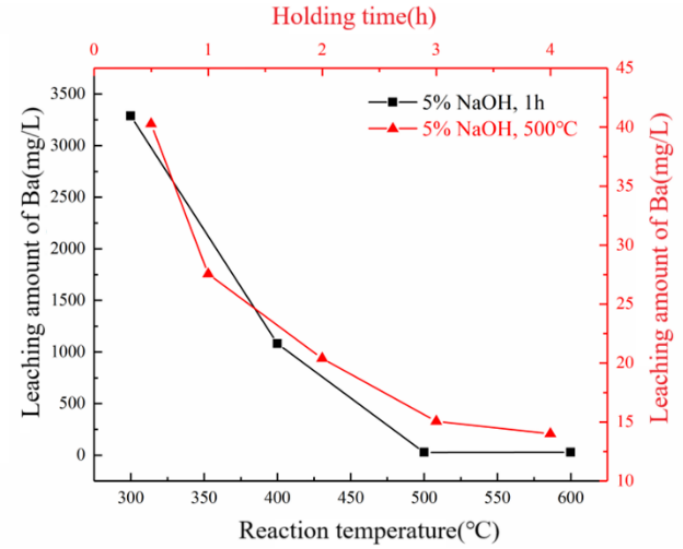


Figure 5. The variation trend of Ba leaching amount affected by three factors

In summary, holding time at 500℃ and 0.5h is a suitable treatment condition, which can effectively cure Ba element, and the leaching results after curing can also meet the national standards.

4. CONCLUSION

In order to realize the "harmless, resource-based and reductive" treatment of oil-based cuttings, the alkaline thermal reaction experiment was carried out in a shale gas well. The curing and treatment of heavy metal Ba in oil-based cuttings were experimentally studied by changing the reaction temperature, the proportion of additives and the holding time. The main conclusions are as follows:

1) The alkaline heat treatment technology has a good curing effect on Ba and can significantly reduce the leaching amount of Ba.

2) The curing effect of Ba was significantly affected by the reaction temperature, the reaction rate could be increased with the increase of temperature, and the reaction degree could be increased with the increase of the addition of NaOH;

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