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Analysis of the Effects of Alkaline Heat Treatment of Heavy Metals from Oil-based Rock Chips on As Elements

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Abstract

This paper focuses on oil-based rock chips produced from a shale gas well in Chuannan, Sichuan, China. Thermal treatment the effects of reaction temperature and NaOH addition ratio on the leaching characteristics of As elements from oil-based rock chips during alkaline thermal treatment physics were studied using NaOH as additive. Experimental results showed that NaOH addition during heat treatment would lead to more leaching of As; Meanwhile, as leached out more and more with increasing reaction temperature and NaOH addition proportion, and after NaOH addition proportion was more than 5%, there would be an obvious cross-over increase. The analysis of experimental results shows that the better treatment conditions for alkaline heat treatment of oil-based rock chips are 5% NaOH addition and a reaction temperature of 500℃.

Keywords

Oil Based Rock; As; Alkaline Heat Treatment; Reaction Temperature; NaOH.

1. Introduction

Oil based rock chips are companion pollutants during shale gas extraction and present a huge pollution to the environment, while with the continuous growth of shale gas industry in China, the production of oil-based rock chips is also increasing, and also becomes one of the main problems limiting shale gas development in China[1]. Therefore, how to prevent the hazards of oil-based rock chips, protect the ecological environment, enhance the capacity of oil-based rock chips disposal, and realize the " harmless, resourced, and abatement " treatment of oil-based rock chips is the focus of scholars at home and abroad[2,3].

There have been many years of research on rock chip treatment technologies for oil machines at home and abroad, and the main treatments are pyrolysis treatment, extraction treatment, biological treatment, backfill treatment, chemical cleaning and other technologies[4-12]. Oil based rock chips after pyrolysis can reduce the oil content to below 0.3%, the residue after pyrolysis can be used as building materials or basis filling and so on; Microwave heating treatment technology, which is also one of pyrolysis, and with the increase of water content, the lower oil content after microwave treatment, and compared with electrical heating, microwave heating is more beneficial to the pyrolysis of petroleum hydrocarbons. The oil phase recovery can reach 99.65% under optimal conditions with supercritical CO2, and the removal efficiency of organic carbon can reach 89.2% in 10s under 500℃ with supercritical water, and studies by Truong Xuan do et al indicate that this type of technology has some economic potential. Tibor et al studied the species of microorganisms under aerobic and hypoxic conditions, and obtained 23 species with good decomposition ability. Also reported that oil removal from rock chips could reach 95.7% after treatment using nanoemulsion cleaning process. These methods mainly remove the oil from oil-based rock chips and have little effect on their heavy metals pollution treatment[13-20].

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However, contaminants vary from region to region, so it leads to different treatment methods. Oil based rock chips from Chuannan region because of its composition, the way of alkaline heat treatment is effective and can effectively solidify the heavy metal pollution in them, however, the situation for the change of As in them is not very clear, so in this paper, alkaline heat treatment is used to treat As in oil-based rock chips, and the change trend and mechanism of As are studied.

2. Experimental Materials and Methods

2.1 Experimental Materials and Instruments

The main experimental materials used were: Deionized water, sodium hydroxide (NaOH), oil-based rock chips. In which deionized water was used as leaching solvent and NaOH as alkaline additive for alkaline thermal reaction, oil-based rock chips were obtained from a shale gas well in Chuannan, Sichuan, China.

Main experimental instruments used were box electric furnace (tn-sx2-12-10n), blast drying oven (bg2-76), horizontal shaker (jc-ggc-12w), energy spectrometer (410-m), SEM (vega3sbu), atomic emission spectrometer (Agilent icp-oes730), X-diffraction analyzer (D8 advance).

2.2 Experimental Approach

In the alkali heat curing treatment, the curing effect of heavy metals is mainly affected by three factors, such as reaction temperature, holding time and NaOH addition ratio, which are changed experimentally to obtain as element curing and leaching characteristics by using orthogonal experiments. The internal incorporation ratio of NaOH in the experiment was 0, 3, 5, 7, 9%, and the reaction temperature was 300, 400, 500, and 600℃, respectively, and the incubation time was 1h.

After alkaline heat treatment, the material composition of the samples was acquired by XRD detection, and the distribution of As elements was analyzed by SEM mapping; The sample leaches were examined for heavy metal leaching by ICP-OES according to the regulations related to the horizontal oscillation method for toxic leaching of solid waste (hj557-2010), and the values were compared with those of the reference standard (50) 3-2007 leaching toxicity identification of hazardous waste identification criteria.

2.3 Oil based Rock Chips Basic Properties

Fig. 1 presents the basic morphologies of the experimental samples of oil-based rock chips. As shown in Fig. 1, the host of oil-based rock is a sticky solid that is greyish black and mainly composed of oil-based drilling fluid, ground rock debris and soil, etc. all oil-based rock chip samples in this paper were obtained from a shale gas well in the region of Argonne Rongxian, and the oil-based rock chip samples had an oil content of 6 ~ 7% and an aqueous content of 5 ~ 8%.

Fig. 1 Initial form of oil base debris

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3. Analysis of Experimental Results

3.1 Effect of Additive Ratio on As Leaching Amount

In alkaline heat treatment, there are three main influencing factors, which are treatment temperature, additive ratio and reaction time

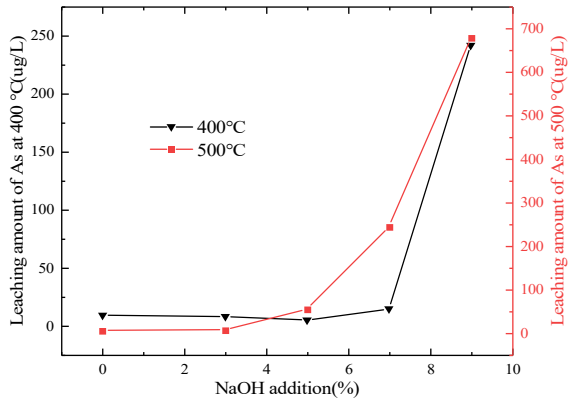

Fig. 2 Variation trend of As leaching amount under different NaOH addition ratio

Fig. 2 is the leaching amount of As changing trend under different NaOH addition proportion conditions. It can be found from the figure that, whether the reaction temperature is 400℃ or 500℃, more NaOH will increase the leaching amount of As and present a negative effect on the treatment of As. At NaOH addition ratios below 5%, As leaching amounts almost unchanged, indicating that NaOH has little role on As at this time; When the NaOH addition proportion was higher than 5%, the leaching amount of as increased rapidly, and this trend was more obvious at 500 ° C, which indicated that the addition of more NaOH was unfavorable to the stabilization of As.

This happens, mainly because of the higher priority of NaOH reacting with other heavy metals, during the treatment, when the NaOH addition is less, NaOH reacts completely with compounds from elements such as Ba and S, but not with As; When the NaOH addition is more than 5%, there will be more NaOH remaining, and these remaining NaOH will go through the reaction of formula (1) with As compound to generate the unstable Na3AsO3. The more NaOH was added, the deeper the extent to which this reaction proceeded.

$$As_2O_3 + 6NaOH = 2Na_3AsO_3 + 3H_2O \quad (1)$$

3.2 Effect of Reaction Temperature on As Leaching Amount

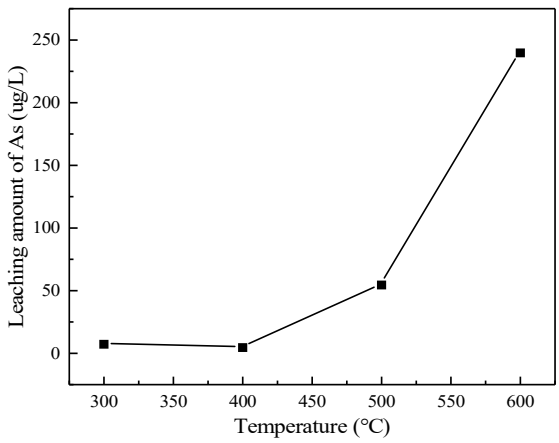

Fig. 3 Variation trend of leaching amount of As under different reaction temperatures

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According to the results of the previous section, there was a significant demarcation in the effect of 5% NaOH addition ratio on As, so the NaOH addition ratio was constant at 5% in the study of reaction temperature and holding time.

Fig. 3 is the change trend of leaching amount of As at different reaction temperatures, from the figure, it can be found that the leaching amount of As is also increasing with the increase of temperature, after exceeding 500 °C, the increase is significantly larger, indicating that the increase of temperature will promote NaOH to react with the compounds of As, and the higher the temperature, the promotion effect is more obvious.

This is mainly because the melting point of NaOH is around 318℃, so at 300℃, NaOH is still in the solid state and cannot provide a liquid reaction environment, instead, the As element will not react with NaOH, so the leaching of As is little, basically consistent with the initial.

Upon continued warming, NaOH gradually becomes liquid, which can not only destroy the crystal structure of compounds, but also provide a liquid reaction environment to make NaOH and other compounds in oil-based rock chips chemically active, can react with NaOH under this condition, the higher the temperature, the more thoroughly liquefaction of NaOH, the stronger the chemical activity will also.

In summary, the addition of 5% NaOH at 500℃ is a suitable treatment condition, under which NaOH, when curing other heavy metal elements, also does not lead to the leaching of As in excessive amounts and generates new fouling.

4. Summary

In order to realize the " harmless, resourced and abatement " treatment of oil-based rock chips, this paper is aimed to develop an experimental study on alkaline thermal reaction generated during the extraction of a shale gas well in Chuannan, Sichuan, China. Experimental studies on the changing trend and mechanism of as during alkaline heat treatment of oil-based rock chips by changing the reaction temperature and additive ratio with NaOH As additives are carried out, and the main conclusions are as follows:

1) Alkaline heat treatment has a negative effect on the leaching of as and will increase the leaching amount of As;

2) When the NaOH addition proportion was lower than 5%, the leaching of As showed a small increase, but the leaching of As increased rapidly after the addition proportion was higher than 5%;

3) The experimental results show that the conditions for better alkaline heat treatment of oil-based rock chips are as follows: 500 ° C, 5% NaOH addition ratio, under which the leaching amount of As does not increase greatly and will not cause secondary pollution;

4) The inadequacy is that the samples as a whole presented strong basicity after treatment, and the pH values of leachates were in the range of 12 ~ 13 at 500℃ with 5% NaOH added ratio, which need further treatment, such as being used as deacidifying agent and so on.

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