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Research article

The study on pyrolysis of oil-based drilling cuttings by microwave and electric heating

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ABSTRACT

In this paper, the following questions were investigated: the proportion of mass loss, the mass fraction of oil, the structure, composition and ultimate analysis of solid residues and gas products. By comparing the treatment effect of using both microwave and electric as the source of heat to dispose the oil-based drilling cuttings (OBDC), the advantages of microwave heating treatment were demonstrated. Meanwhile, the composition of liquid products by microwave pyrolysis was analyzed. The results show that the microwave heating is better than electric heating and the former can promote the pyrolysis of petroleum hydrocarbons. The results of component analysis of the liquid products from OBDC by microwave pyrolysis show that C12 ~ C20 components pyrolyze at 500 °C. At the same time, a mass of C21 ~ C24 components volatilize. At the temperature above 500 °C, the thermal cracking reactions of > C25 components occur and a maximum content of paraffin in liquid products is obtained. As the temperature increases, the components obtained by pyrolysis become more and more complex.

1. Introduction

Owing to the increase of energy consumption, researches on the exploitation of shale gas has become the center of development strategies in various countries. As a result, the disposal of oil-based drilling cuttings (OBDC) was of great concern. During the exploration and extraction of oil and natural gas, rock fragments carried by the drilling fluid to the surface are referred to OBDC. The OBDC are collected and stored in a tank or pit for further treatment or management after additional mechanical processes. In recent years, there was a growing demand for environmental requirements, the previous treatment methods for OBDC, such as bioremediation (Guerra et al., 2018; Paladino et al., 2016) and solvent extraction (Goodarznia and Esmaeilzadeh, 2006), has gradually been eliminated because of the difficulties of transporting the OBDC to land.

Pyrolysis was considered one of the most promising approaches for the treatment solid wastes, and the tendency has been investigated on many different types of solid wastes including algae, biomass, municipal solid wastes and sewage sludge (Domínguez et al., 2005, 2006; Gedam and Regupathi, 2012; Hu et al., 2012). It can prevent the generation of secondary pollutants effectively because of its inert environment and low temperature. Furthermore, the solid residues, gas and liquid products produced by pyrolysis have a wide range of applications. For example, the gas and liquid products can be used as fuels, and solid residues can be used as building materials and cheap adsorbents for contaminants (Lu et al., 2012). The most common methods for the pyrolysis process in treating solid wastes are microwave heating and electric heating (Beneroso et al., 2015; Hossain et al., 2011; Robinson et al., 2008b; Shang et al., 2007). Zhao et al. (2012) reported the effect of temperature on the microwave pyrolysis of biomass (wheat straw). LU Tao et al. (Lu et al., 2012) reported the influence of pyrolysis temperature on biochar, liquid and gas fractions in the pyrolysis process of sewage sludge. Lin et al. (2012) developed a microwave reactor to produce bio-oil from sewage sludge. Domınguez et al. (2003) compared the bio-oil composition produced from microwave-assisted and conventional pyrolysis. A pilot study on removal of oil content from OBDC demonstrated that oil-contaminated cuttings can be remediated to below 1% (w/w) using a high power multimode cavity (Shang et al., 2006b).

Electric heating is a traditional mode used in heating process, and heat is transferred from the surface to the center of the material by conduction, convection, and radiation. The method depends on the thermal conductivity of the material and the heat transfer efficiency in the system (Motasemi and Afzal, 2013). On the contrary, microwave heating has different heat transfer and mass transfer mechanism compared with electric heating, which can generate thermal energy by the

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interaction of electromagnetic fields with molecules in the material. Because of the penetrability and selectivity, the microwave can directly act on each individual unit inside the material (Robinson et al., 2008a). The method depends on the characteristic of materials such as dielectric properties, and so on. As far as the interaction with microwaves, materials can be classified into three generic classifications (Chandrasekaran et al., 2012; Shang et al., 2006b):

(1) Absorbers or high dielectric loss materials—microwave energy is absorbed on account of the dielectric loss factor;
(2) Opaque or electrical conductors—microwaves are reflected and do not penetrate.
(3) Transparent or low dielectric loss materials—microwaves are transmitted through the material with little attenuation.

A research shows that the treatment time of microwave heating can usually be reduced to 1% of the time required to use in electric heating method (Meredith, 1999).

In this work, the effects of the treatment of OBDC using electric heating and microwave heating were examined and compared. Meanwhile, the components of the liquid products from OBDC by microwave pyrolysis were collected and analyzed. The study can provide a theoretical basis for the microwave pyrolysis and the resource utilization of OBDC.

2. Materials and methods

2.1. Experimental apparatus

The schematic diagram of the microwave pyrolysis system during this study was shown in Fig. 1. The microwave reactor was manufactured by Hunan Changyi Microwave Technology Co. LTD (Changsha, China). A 0–1.4 kW microwave generator (2.45 GHz) was used for microwave oven. The pyrolysis temperature was measured by a high temperature K-type thermocouple (Φ3 mm). The OBDC sample was placed in a railboat with a hole at the end within a cylindrical quartz glass reactor (the thick pipe, Φ40 mm × Φ34 mm × 363 mm; the thin pipe, Φ14 mm × Φ10 mm × 220 mm), insuring that the thermocouples were inserted into the material. For each experiment, approximately 20 g of OBDC was used, and N_2 was continuously supplied at a flow rate of about 200 mL min^{-1} to maintain the inert environment as well as to sweep the vapor out of the reactor. Microwave power (700 W) was kept constant for each test and the treatment time was 20 min. The microwave reactor was equipped with an automatic temperature/power control to maintain the desired reaction temperature. The system was purged with N_2 for 15 min at a flow rate of 150 mL min^{-1} before pyrolysis reaction to keep an O_2-free environment. The vapor generated out of the reactor passed through two consecutive ice-water cooling condensers (0 °C) to collect the condensed oil and water. The non-condensable gas was then collected with a gas-collecting bag. The liquid and gas products were analyzed by a gas chromatography and gas chromatography-mass spectrometer. The process and apparatus of electric heating were similar to those shown in Fig. 1, except the microwave oven was changed to electric furnace and a heating rate of 5 °C·min^{-1} in electric heating process. The electric heating reactor (power rating – 1000 W) was manufactured by Anhui Best Equipment Technology Co. LTD (Hefei, China).

2.2. Materials

The oil-based drilling cuttings (OBDC) were collected from the well (YS112) drilling in Sichuan province, China. In order to reduce the impact of differences in particle size distribution, firstly, OBDC were crushed using a micromill (Portable high-speed mill, QE-100). The average diameter was measured by a Laser Particle Sizer (NanoZS90new, UK) about 14.18 μm (Table S1). The reservoir cuttings used in this work are rock fragments from onshore well drilling with initial content of 3.34% oil. The properties of the OBDC were listed in Table S2. The results indicate that the original OBDC contain more than 85% of ash, which greatly limits the resource utilization of OBDC solid residues. In addition, the original OBDC consist of 4.59% moisture, 13.02% volatile and 1.57% fix carbon.

2.3. Material characterization

The elemental analysis of carbon (C), hydrogen (H), nitrogen (N), sulfur (S) and oxygen (O) of OBDC was performed on a CHNS/O elemental analyzer (Elementar Vario ELIII, Germany) according to the Chinese standard (JY/T 017-1996). The original oil (O-oil) and water content was investigated by the method of the Soxhlet extraction with carbon tetrachloride and azeotropic distillation with toluene according to the Chinese standard (GB/T 260-2016 and GB/T 16488-1996 respectively). The ashes, volatile and fixed carbon were determined according to the American Society of Testing Materials (ASTM) standard. The structure and composition of solid residues were characterized by Scanning Electron Microscopy (SEM, HITACHI-S-4800, Japan), Fourier Transform Infrared Spectrometer (FT-IR, Nicolet Avatar 370, USA) and X-Ray Diffraction (XRD, Bruker D8-Advance diffractometer with Cu Kα).

The gas products were analyzed by a gas chromatography (GC,

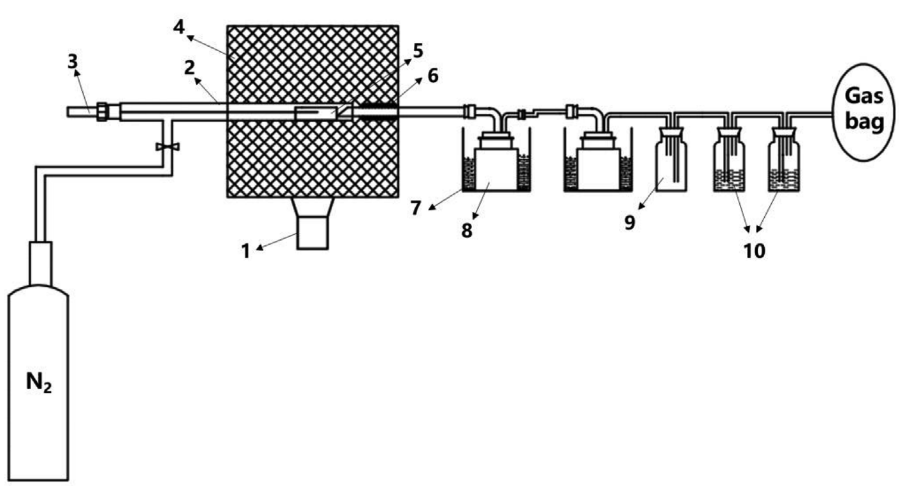

Fig. 1. The schematic diagram of pyrolysis process of OBDC by microwave. Keys: (1) Microwave generator; (2) Quartz tube; (3) thermocouples; (4) Insulating brick; (5) railboat; (6) The coil of electric heating; (7) Ice-water bath; (8) Pyrolysis oil collector; (9) Safety bottle; (10) The treatment of tail gas (containing NaOH solution).