



Study on the characteristics of microwave pyrolysis of high-ash sludge, including the products, yields, and energy recovery efficiencies

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ABSTRACT

The pyrolysis residues of high-ash sludge are rich in the metallic oxides of silicon (Si), iron (Fe), and aluminum (Al), enabling high-ash sludge to achieve complete pyrolysis without the addition of wave-absorbing materials. Therefore, these residues offer obvious advantages when compared with traditional sludge when pyrolyzed under microwave irradiation. The differences in final pyrolysis temperatures caused by different microwave powers have significant effects on the characteristics of pyrolysis products. By increasing the microwave power, the lower heating value of bio-gas increased with the increase in syngas output, the lower heating value of bio-oil first increased and then decreased with changes in hydrocarbon yields, and the specific surface area of bio-char first increased and then decreased; however, the adsorption capacities of heavy metals, such as copper (Cu), chromium (Cr), and nickel (Ni) increased gradually. The yields of bio-gas and bio-oil increased from 10.01% to 2.98%–14.02% and 3.52%, respectively, as the input power increased from 700 W to 1300 W. Thereby, energy recovery efficiency reached the maximum at 5.15% with a microwave power of 1300 W.

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1. Introduction

Since Menéndez et al. first reported the study of treating urban sludge with microwave technology, extensive studies have been conducted on the principle of microwave pyrolysis of sludge [1]; main influence factors, including microwave power [2], wave-absorbing materials [3], and catalysts [4]; characteristics of the products; efficiency of pyrolysis [5–7]; main element migration conversion rules [8]; and heating mechanisms [9–11]. Compared with traditional sludge pyrolysis technology, results have shown that microwave pyrolysis technology had obvious advantages in terms of increasing resource recovery efficiency by means of copyrolysis with organic raw materials, such as crop stalks, nut shells, and fruit peelings; increasing the yields of biofuels, such as bio-oil and bio-gas; and strengthening the heavy metal

solidification to reduce environmental pollution risks [12]. It has been widely believed that microwave pyrolysis technology holds the greatest potential for development of sludge treatment in the future [13–15].

Even with these advantages, however, problems such as the resource and energy consumption that occurs in microwave pyrolysis should not be overlooked. Such problems inevitably limit the application of microwave pyrolysis of sludge.

- (1) Resource consumption resulted from adding wave-absorbing materials or catalysts: extensive studies show that adding wave-absorbing materials, such as silicon carbide (SiC), graphite, and activated carbon, was the essential condition for realizing high-temperature pyrolysis under microwave irradiation [16–18]. Meanwhile, to improve the yields, qualities, and safety of the bio-gas and bio-oil generated, research gradually focused on studying the addition of catalysts, such as iron (III) oxide (Fe₂O₃), calcium oxide (CaO), and aluminum oxide (Al₂O₃), in the microwave pyrolysis. Adding catalysts could increase the heating value of bio-oil

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and the yields of syngas by about 10% and 20%, respectively [19,20]. The high price of the wave-absorbing materials and catalysts, however, significantly increased the resource consumption and total costs of microwave pyrolysis of the sludge. In addition, the environmental pollution that occurred during production and the discharge of these materials could not be overlooked. The reasonable control of resource consumption that occurs during microwave pyrolysis is essential to realizing the wide application of this technology.

- (2) Energy consumption occurs during the microwave energy input: The input power of microwaves directly affected the conversion of microwave energy to thermal energy, thus influencing the rising temperature and weight loss of sludge. The microwave energy inputs required by the sludge pyrolysis were calculated and controlled based on the physico-chemical properties of the materials and the initial and final temperatures of pyrolysis. The excessively high input powers could lead to local overheating and could reduce the utilization efficiency of energy sources, while low input powers could affect the yields and qualities. To date, most studies have focused on the characteristics and yields of pyrolysis products under fixed microwave powers [21–24], but product outputs at different microwave powers have been less studied often. Thus, it remains unknown whether microwave pyrolysis technology could realize efficient resource recovery under low energy consumption.

It remains difficult to reduce the resource and energy consumption in microwave pyrolysis. Breulmann et al. [25] pointed out that pyrolysis residues produced from the microwave pyrolysis of sludge could participate in situ in the pyrolysis as a wave-absorbing material, which offered a good option to reduce resource consumption. In recent years, researchers have tried to use these pyrolysis residues as wave-absorbing materials to participate in the pyrolysis of sludge [21,26], and results have shown that the pyrolysis residues could function as traditional absorbing materials to realize the complete pyrolysis of sludge at high temperature. Until now, however, it remains unknown why the pyrolysis residues can realize the absorption of microwave energy and how about the energy recovery efficiency occurs with pyrolysis residues as the wave-absorbing material is still unknown.

In terms of the wave-absorbing materials required for the microwave pyrolysis of sludge, the substances that have wave-absorbing functions include the oxides and crystal compounds, such as SiC, silicon dioxide (SiO_2), and Fe_2O_3 , of such elements as Si, Fe, and zinc (Zn), which also could change the migration and conversion of the organic elements in the sludge and thus could realize the directional catalysis in the microwave pyrolysis of sludge. Therefore, we could infer that the high-temperature pyrolysis of sludge could be realized without the addition of wave-absorbing materials and catalysts, if these metal elements are included. Moreover, some related studies on the microwave pyrolysis of coal also indicated that rich components, such as SiO_2 , Al_2O_3 , FeO, Fe_2O_3 , and CaO, could function as wave-absorbing materials and catalysts [17,27]. Thus, it appears that the microwave pyrolysis of high-ash sludge has comparative advantages against traditional sludge with a high content of organic matter in terms of energy saving and resource consumption. If this hypothesis is tenable, then co-pyrolysis could be used for high-ash sludge and other environmental wastes with a high content of organic matter, and the energy input and output of the pyrolysis system could be optimized by regulating input power. Therefore, low energy consumption and efficient reclamation of resources could be realized at the same time. Although the study of product distributions, yields, and

energy recovery efficiencies of the microwave pyrolysis of high-ash sludge has significant importance, no study in this field has been reported to date.

This paper uses the dewatered high-ash sludge from an industrial sewage plant in Shenzhen City as an experimental material. We first compared the yields of products and the energy recovery efficiency of high-ash sludge that had been pyrolyzed with different wave-absorbing materials to reveal the advantages of microwave pyrolysis of high-ash sludge in resource and energy saving. Then, without adding wave-absorbing materials, we studied the change rules of the product distributions, the yields, and the energy recovery efficiencies of the microwave pyrolysis of high-ash sludge to realize an efficient reclamation of organic resources in high-ash sludge with low energy consumption. This paper could (1) address gaps in studies on the rules of the microwave pyrolysis of high-ash sludge; (2) identify the characteristics of the resulting products, creating conditions for the low energy consumption and efficient co-pyrolysis of high-ash sludge and traditional domestic sludge, garbage, fruit peelings, and other wastes; and (3) provide a theoretical basis for reducing the total costs of the microwave pyrolysis process.

2. Materials and methods

2.1. Experimental materials

To study the microwave-absorbing properties of the pyrolysis residue generated from the microwave pyrolysis as well as the characteristics and energy recovery efficiencies of the pyrolysis of high-ash sludge, we used dewatered high-ash sludge collected from an industrial sewage plant in Shenzhen City as the feedstock. The main characteristics are listed in Table 1. The sludge had a moisture content of 88.10 wt %, an ash content of 61.88 wt %, an organic substance content of 38.12 wt %, and a lower heating value (LHV) of 2.76 ± 0.12 MJ/kg. The test results for ash analysis indicated that the contents of Si, Fe, and Al were more than 10.10 wt % and all existed in the form of oxides.

2.2. Experimental methods

We used the same devices for the microwave pyrolysis of high-ash sludge as those illustrated in Ma et al. [28], including a thermocouple thermometer (temperature range: 0–1700 °C) and an electrical scales (accuracy at 0.001 g) used for online measurement of the materials' temperatures and masses in the quartz tubes. We first used 5 g SiC, pyrolysis residue (produced from pyrolysis of high-ash sludge), and graphite as the wave-absorbing materials and mixed these materials with 25 g of high-ash sludge to form a mixed sample of 30 g. We then placed the samples into quartz tubes and maintained the microwave power at 1000 W to study the characteristics and yields of the microwave pyrolysis of high-ash sludge with the addition of different wave-absorbing materials. Thus, we explored the advantages of microwave pyrolysis of high-ash sludge compared with traditional sludge. Then, we put 30 g of high-ash sludge without any wave-absorbing materials into quartz tubes and adjusted the input power to four different levels of 700 W, 1000 W, 1300 W, and 1600 W to study the change rules of the characteristics of the product distributions, yields, and energy efficiency of the microwave pyrolysis of high-ash sludge. To guarantee an inert-gas environment, we pumped high-purity nitrogen into the microwave cavity for 20 min before the experiment, controlled the flow rate at 200 mL/min, and then adjusted the flow rate to 10 mL/min after the feeding of microwave to the sludge. The high-temperature gas produced by the pyrolysis reaction entered a condensing device, where part of the gas condensed into an oily

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