



Research Paper

Thermal behavior research for co-combustion of furfural residue and oil shale semi-coke



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HIGHLIGHTS

- Combustion behavior of oil shale semi-coke and furfural residue was studied.
- The effect of heating rate and mixing ratio were researched.
- The mutual effect of blends occurred in the process of co-combustion.
- The kinetics parameters were calculated by Coats-Redfern method.

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ABSTRACT

The thermal behavior of Longkou oil shale semi-coke, furfural residue and their blends was researched in this paper. The experiment was carried out using TG-FTIR to research the combustion mechanism, under different heating rate (10, 20, 40 and 80 °C/min) and simulation of air condition (80% nitrogen: 20% oxygen). The oil shale semi-coke was blended with furfural residue in proportions from 20% to 100%. Two following factors were studied: heating rate and mixing ratio. The combustion behavior of semi-coke can be promoted furfural residue. And the synergistic effect was calculated by means of interaction coefficient f and the Relative Error of Mean Square root. The combustion of two samples can be divided into two stages, and three stages of co-combustion. Synergy occurred in every stage. At the same time, the co-combustion kinetics parameters were calculated by the Coats-Redfern method. The activation energy of the mixtures first decreased and then increased with the degree of reaction.

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1. Introduction

In China, oil shale is still regarded as an essential supplement for limited petroleum resources [1]. Semi-coke generated from the process of oil shale retorting, which still contains a certain calorific value, is always abandoned. Generally, every ton of shale oil production produces about 10–30 tons of semi-coke [2]. So much semi-coke discharge not merely occupies abundant land, but does harm to environment [3], from which the leachate contains sulfide, hydrocarbons, poly-cyclic aromatic hydro-carbon [4], and trace amounts of other toxic elements.

Until now no large-scale utilization of these solid wastes has been found in China. Some researchers suggested burning petroclastic shale, retorting and excess gas to generate electricity [5,6]. Opik et al. [7] proposed that mixing some oil shale into semi-coke was necessary to supplement insufficient heat value from

semi-coke for stable combustion. Arro et al. [8] compared different combustion modes and proposed to burn the mixture of semi-coke and oil shale by means of fluidized bed combustion. Kaijuvee et al. [9] studied the adsorption property of sulfur of oil shale, semi-coke and their mixture when burning in the fluidized bed, in which he concluded the semi-coke can absolutely adsorb the SO₂ generated. Qin et al. [10] studied the co-combustion kinetics of shale oil sludge and semi-coke, and found the oil shale sludge improved the combustion of semi-coke.

Furfural residue, generated after extracting furfural from corn-cob, is a kind of biomass waste. There generates about 12–15 tons of residue per ton of furfural production. Furfural residue is rich in cellulose, hemicellulose and lignin, which own great recycling value [11]. However, its high salt content may pollute the atmosphere, soil and river if abandoned directly. Therefore, effective measures should be taken to eliminate these detrimental influence in the furfural industry.

It is noteworthy that the two solid wastes have a certain calorific value, which are available for energy utilization by putting them

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into boiler. Semi-coke of oil shale is hard to ignite solely because of a lower calorific value contained. While it can be solved by mixing the two materials because furfural residue contains much calorific value close to bituminous coal, which supplements insufficient heat of semi-coke. In addition, co-combustion of furfural residue and oil shale semi-coke may hopefully become components into non-hazardous. Thus semi-coke of oil shale and furfural residue being made energy use may be an ideal solution.

Till now few reports have been found for co-combustion research of semi-coke and furfural residue. While a number of studies have been conducted for co-combustion of other fuels. Liu et al. [12] studied thermal behavior and the synergy of oil shale semi-coke with torrefied cornstalk, and concluded that co-combustion increased with greater degree of heat-treated cornstalk, and interpretation of the MR curve indicated that the influence of the torrefied cornstalks on the combustion process is observed mainly during the second phase [13]. Gayan et al. [14] found that blending biomass improved combustion efficiency when burning coal in a circulating fluidized bed. Sahu et al. [15] selected different biomass carbo-coal mixing with semi-coke to research combustion kinetics through DSC-TGA experiment. The results showed the burnout rate increased with burning time for combustion of coal mixed with less than 50% biomass. Varol et al. [16] found the ignition temperature of biomass and low grade lignite was close to pure biomass when burning in a thermogravimetric analyzer. Wang et al. [17] found the temperatures of ignition and burnout reduced with the increasing rice straw proportion.

In actual engineering project, combustion of biomass is a low cost low risk of renewable energy utilization way. It's significant for complementing the shortage of fossil fuels by the burning of these two abandoned residues, and reducing the emissions of traditional pollutant (SO_2 , NO_x) as well as greenhouse gas (CO_2 , CH_4). The achievement of this study may provide reference for renewable energy application. Also it is meaningful for conventional fossil fuel consumption in energy production in China.

2. Experimental section

The experiments were performed on a thermogravimetric Analysis TGA/DSC1 (Mettler-Toledo, Switzerland) and Fourier Transform Infrared Spectrometer (Nicolet IS10, Thermo Scientific, United State). 10–20 mg of each sample is prepared in the tests. Four heating rates were arranged, i.e. 10, 20, 40, 80 °C/min, and the reaction temperature at 50–950 °C. The gas atmosphere was synthetic air with 50 mL/min ($\text{O}_2:\text{N}_2 = 1:4$). The spectral detection ranged between 4000 and 400 cm^{-1} , with 4 cm^{-1} of resolution, and 20 kHz of 200 cycles of scanning rate.

The semi-coke was made by retorting Longkou oil shale. Furfural residue was selected from a furfural factory located in Shuangyang District, Changchun city. The samples were ground into fine particle (<0.2 mm). Owing to high moisture contained in the furfural residue, a drying process was needed. The furfural residue was blended with semi-coke in certain proportions. The data in detail are listed in Table 1 where FR and SC are respectively the abbreviations of furfural residue and semi-coke.

Table 2 shows the fuel property of two fuels, containing proximate, ultimate analyses and heat value. There are some differences between the two materials. The volatile contained in semi-coke is

less while abundant in furfural residue. On the contrary, the ash content of semi-coke is much higher than that of furfural residue. The ash and volatile content of the two fuels forms complementary trend, so it is possible to promote properties for each other during their combustion.

3. Results and discussion

3.1. Combustion of pure samples

The curves of combustion for furfural residue and oil shale semi-coke under a heating rate of 20 °C/min are shown in Fig. 1, where Fig. 1a and b are respectively TG-DTG curves of semi-coke and furfural residue.

Fig. 1a shows the combustion of semi-coke can be divided into two stages, thermal decomposition of volatile (360–655 °C) and fixed carbon burnout (655–810 °C). From the figure one can see the sample weightlessness is small because of low content of surplus volatile and fixed carbon for semi-coke. The combustion mainly concentrates on release and thermal decomposition of volatile at 360–655 °C in the first stage, with only 16% of weightlessness. While the second stage mainly shows smaller weight loss than the first with only 8% of weightlessness at 655–810 °C, in which a peak occurs at 760 °C.

Fig. 1b shows two stages are also contained in the whole combustion process of furfural residue, namely burning of volatile and fixed burning out of carbon. The first stage occurs at 260–370 °C, where the weight loss reaches 49%. And 43% of weightlessness takes place at the second stage (370–680 °C). Unlike semi-coke, furfural residue contains abundant components materials volatile and fixed carbon, which lead to so much range variation in mass of sample. It is also deduced that furfural residue behaves superior to semi-coke. From the DTG curve of furfural residue, it is shown two peaks distributed in the process. It's noteworthy that the amplitude of first one is larger than the latter, while the first peak width smaller than the latter. This phenomenon just confirmed the judgment of two stages distribution.

Generally, fluidized bed combustion not only need sufficient burning, but a substantial non-combustible solid materials save considerable heat to maintain bed temperature. The ash of semi-coke may possess thermal storage capacity. Because of the disparate behaviors between the two samples during combustion, there might be possible to realize favoring combustion characteristics to each other when mixes them together.

3.2. Effect of mixing ratio

Fig. 2 shows TG-DTG curves of four kinds of blended samples under the heating rate of 20 °C/min. From the figure b, one can see that there are three peaks during the mixture co-combustion process, therefore the co-combustion can be divided into three distinct phases: The combustion stage of volatile in the furfural residue, releasing and thermal decomposition of volatile of semi-coke and burning of fixed carbon of furfural residue, and, burning of fixed carbon in the semi-coke. With the increase of furfural residue proportion, the weightlessness rate of samples significantly increased in the TG curve; and the DTG curve shows the weightlessness rate increase in every stage, especially in the first two

Table 1
Proportion of sample mixtures.

Serial number	S ₁	S ₂	S ₃	S ₄	S ₅	S ₆
FR:SC	0:10	2:8	4:6	6:4	8:2	10:0

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