



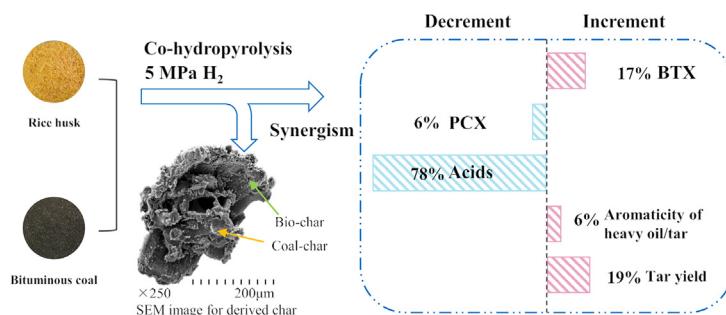
Comparative investigation of rice husk, thermoplastic bituminous coal and their blends in production of value-added gaseous and liquid products during hydropyrolysis/co-hydropyrolysis

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GRAPHICAL ABSTRACT



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ABSTRACT

The hydropyrolysis/co-hydrogenation of rice husk (RH) and thermoplastic bituminous coal (BC) was carried out using a fixed-bed reactor to investigate the effects of atmosphere and hydrogen pressure on product distributions. RH produced more carbon oxides, phenolics and acids. BC yielded more methane and BTX (benzene, toluene and xylene) during hydropyrolysis. Compared with hydropyrolysis of RH, the co-hydrogenation promoted the higher heating value of gaseous product and the yield of BTX by 19% and 57% respectively, while it reduced the yield of corrosive acids by 89% under 5 MPa H₂. The yields of methane, BTX and phenolics during co-hydrogenation were 1.5, 6.4 and 4.0 times as much as those obtained during co-pyrolysis under 5 MPa. The co-hydrogenation reformed the structure of heavy oil/tar, benefiting the development of aromaticity. High hydrogen pressure synergistically reduced yields of char and acids, and enhanced yields of tar and light aromatics via the secondary reactions.

1. Introduction

A worldwide rapid economic and social development has led to massive production and energy consumption. It arises problems like depletion of resources, environmental pollution, ecological damage etc. Recently, the coal-dominated power generation suffers a global

pressure to reduce greenhouse gas emissions (Liang et al., 2013). Therefore, it is urgent to build a low-carbon, clean and efficient modern energy system by revolutionizing the energy production and consumption process. In China, according to Chinese Academy of Engineering, the consumption ratio of coal, non-fossil energy, oil and gas will adjust from 6:1.5:2.5 (before 2020) to 4:3:3 (before 2050). The

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diversity of energy sources has thus become an important method to balance the energy demand with the fragile ecological environment.

The clean utilization of abundant and broadly distributed non-edible lignocellulosic biomass, which is an renewable and CO₂-free green resource, has been widely investigated during last decades (Gaurav et al., 2017; Ren et al., 2016). However, changes of seasons and geographical environment affect the availability of biomass. The co-pyrolysis (co-Py) of biomass and coal can manage seasonal fluctuations in biomass availability and increase energy density of feedstock. It exerts the advantages of biomass and coal simultaneously to produce liquid chemicals and gaseous fuels.

The early report of biomass and coal co-Py claimed no interdependence between coal and biomass (Klose and Stuke, 1993). Since then, there had been substantial efforts to investigate co-Py/co-gasification and also trials were made to elucidate the probably existing synergistic effect (Guo and Bi, 2015; Wei et al., 2017; Wu et al., 2018). But the work is exceedingly difficult because of the variety of coal/biomass types and the complexity of fuel constituents. In addition, due to the adverse properties of conventional pyrolysis bio-oil, namely low heat value, high intrinsic acidity, high moisture content, instability and corrosiveness, the utilization of derived crude oil is limited (Zhang et al., 2007). Other drawbacks of conventional Py/co-Py are the adverse effects caused by heavy tar formation on the pyrolytic process, which lead to the operational problems such as pipe blocking, condensation, and tar aerosol formation.

Recently, the biomass hydropyrolysis (HyPy), in which hydrogen is used to preferably produce light aromatics and methane, has received researchers' attention (Dayton et al., 2016; Resende, 2016; Zheng et al., 2017). Unlike conventional pyrolysis under inert atmosphere, this technique has high selectivity towards final products. Coal HyPy had been vastly investigated for production of substitute/synthetic natural gas (SNG) and valuable light aromatics which effectively alleviate the SNG production cost (Kopyscinski et al., 2010). Differing from conventional pyrolysis, one important feature of HyPy is the increased carbon utilization of volatiles and the promoted formation of methane and high-quality tar products. However, the higher heating value (HHV) of gases obtained from biomass is much lower than that produced from coal under the same HyPy condition. Meanwhile, substantial corrosive acids would derive from biomass unless catalysts or intensified hydrocracking working conditions were implemented (Zheng et al., 2017). It will increase production costs and lower thermal efficiency. Hence, the study is devoted to exploring co-hydrogenation (co-PyHy) of biomass and a high-volatile bituminous coal. Co-HyPy is expected to improve the selective formation of value-added products via the synergistic effects.

So far, co-PyHy has been only involved in co-utilization of coal/synthetic polymer mixture (Sharypov et al., 2007; Yasuda et al., 2014) and biomass/synthetic polymer mixture (Pinto et al., 2016; Sharypov et al., 2006). Yasuda et al. (2014) implemented co-HyPy of coal/polyethylene (PE) mixture using a batch reactor at 800 °C under 7.1 MPa hydrogen pressure. Results showed that methane yields of mixtures were higher than those calculated from the pyrolysis results of individual fuels. Sharypov et al. (2007) observed the synergistic behaviour from co-HyPy of lignite/polyolefinic plastic using an autoclave. In another work, Sharypov et al. (2006) reported a higher carbon conversion attained by co-HyPy of pine wood and synthetic polymers. Pinto et al. (2016) observed higher yields of gas and liquid by co-HyPy of rice crop wastes and PE. To the best of our knowledge, there are seldom any literature reports pertinent to co-HyPy of biomass and coal. Little is known about compositions of derived oil/tar. In the context, the aim of this work is to investigate the co-HyPy behaviour of rice husk and thermoplastic bituminous coal. Particular attention is paid to the impact of gas atmosphere and hydrogen pressure on quality of derived products and synergies. The study would provide valuable knowledge that can be used to design and operate co-HyPy conversion system of biomass and coal blend.

Table 1

Proximate and ultimate analyses and metallic elements results of rice husk and bituminous coal.

Sample	Rice husk (RH)	Bituminous coal (BC)
<i>Proximate analysis (wt%, db)</i>		
Ash	10.0	16.0
Volatile matter	73.7	29.0
Fixed carbon	16.3	55.0
<i>Ultimate analysis (wt%, daf)</i>		
Carbon	51.2	85.3
Hydrogen	6.2	5.2
Nitrogen	0.4	1.5
Sulfur	0.1	2.8
Oxygen ^a	42.1	5.2
H/C molar ratio	1.45	0.73
O/C molar ratio	0.62	0.05
<i>Metallic elements (wt%, db)^b</i>		
Potassium (K)/ws-ie K	0.42/0.33	0.05/0.00
Calcium (Ca)/ws-ie Ca	0.08/0.09	2.01/2.08
Magnesium (Mg)	0.06	0.15
Silicon (Si)	3.74	2.70
Aluminum (Al)	0.01	2.00
Iron (Fe)/ws-ie Fe	0.02/0.002	1.26/0.0003

^a By difference.

^b The data before forward slash are determined by X-ray fluorescence (XRF), those data after forward slash are obtained by chemical fractionation analysis (ws-ie K/Ca, water soluble-ion exchangeable K/Ca content).

2. Materials and methods

2.1. Samples

The biomass used in this work was supplied by a mill factory in Zhejiang province, China. It was ground and sieved to the particle size range of 0.15–0.45 mm. The componential analysis of dried rice husk (RH) contained 39.8% cellulose, 12.3% hemicellulose, 29.6% lignin and 12.0% neutral detergent solute. A bituminous coal (BC) from Dawanggou district, Sichuan Province, China was used to co-pyrolyze with RH. The coal sample was ground and sieved to the particle size of less than 0.15 mm. Table 1 shows the proximate and ultimate analyses and the contents of the metallic elements in two fuels. RH is enriched with potassium and BC contains high calcium and iron content. The blend of RH and BC was prepared by gently mixing two fuels at a mass ratio of 1:1 (wt%, db) using mortar and pestle for 30 min manually.

2.2. Hydrogenation/co-hydrogenation

HyPy/co-HyPy was carried out in a pressurized two-stage fixed-bed reactor. The schematic diagram, typical longitudinal temperature profiles of the vertical reactor, and detailed operation procedures were described elsewhere (Zhang et al., 2015b, 2017). In this study, the upper zone of reactor was held at 500 °C in all experiments to reduce volatile condensation on the reactor wall. The lower zone of reactor (HyPy zone) was heated up from ambient temperature to 700 °C at a rate of 15 °C min⁻¹ for no holding time at the final temperature. For comparison, similar experiments were conducted under nitrogen atmosphere.

2.3. Analysis of raw fuel/char

Ash compositions of raw fuels were measured by X-ray fluorescence analysis (XRF-1800). Active potassium and calcium in raw fuels were determined by a chemical fractionation analysis (Chen et al., 2016). The morphology and element composition of the surface of char particles were determined using Hitachi SU-1510 scanning electron microscopy coupled with energy disperse spectroscopy (SEM-EDS).

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