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Research article

Investigation of waste biomass co-pyrolysis with petroleum sludge using a response surface methodology

Guangji Hu [a, b], Jianbing Li [a, b, *], Xinying Zhang [c], Yubao Li [a]

[a] WZU-UNBC Joint Research Institute of Ecology and Environment, Wenzhou University (WZU), Wenzhou, Zhejiang Province, 325035, PR China
[b] Environmental Engineering Program, University of Northern British Columbia (UNBC), Prince George, British Columbia, V2N 4Z9, Canada
[c] College of Environmental and Chemical Engineering, Shanghai University, Shanghai, 200444, PR China

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ABSTRACT

The treatment of waste biomass (sawdust) through co-pyrolysis with refinery oily sludge was carried out in a fixed-bed reactor. Response surface method was applied to evaluate the main and interaction effects of three experimental factors (sawdust percentage in feedstock, temperature, and heating rate) on pyrolysis oil and char yields. It was found that the oil and char yields increased with sawdust percentage in feedstock. The interaction between heating rate and sawdust percentage as well as between heating rate and temperature was significant on the pyrolysis oil yield. The higher heating value of oil originated from sawdust during co-pyrolysis at a sawdust/oily sludge ratio of 3:1 increased by 5 MJ/kg as compared to that during sawdust pyrolysis alone, indicating a synergistic effect of co-pyrolysis. As a result, petroleum sludge can be used as an effective additive in the pyrolysis of waste biomass for improving its energy recovery.

1. Introduction

The petroleum industry generates a considerable amount of oily sludge during various processes such as crude oil exploration, transportation, storage, and refining (Hu et al., 2013). This type of sludge is a complex and stable emulsion of various petroleum hydrocarbons (PHCs), water, solid particles, and metals (Hu et al., 2015). It has been classified as a hazardous waste in many countries and thus needs effective treatment. Traditional methods for oily sludge treatment such as landfilling have been challenged by stringent regulations. In recent years, the recycling of energy from such oil-rich waste has received increasing interests, and a variety of methods have been developed (Hu et al., 2013; Jasmine and Mukherji, 2015). Among them, pyrolysis represents an effective thermo-chemical conversion process for both waste disposal and energy recovery (Liu et al., 2009; Qin et al., 2015). For example, Moltó et al. (2013) examined the pyrolysis of two different sludge wastes (i.e., petrochemical sludge and biological sludge) and found that the gaseous products can be significantly affected by the heating rate, oxygen content, contact time, and the nature of sludge. Conesa et al. (2014) investigated the pyrolysis of petrochemical sludge and observed an increased yield of liquid oil (i.e., from 30 to 50%) due to increasing treatment temperature (i.e., from 350 to 530 °C).

In addition to the application to oily sludge waste treatment, many pyrolysis studies have focused on oil production from lignocellulosic biomasses because they are considered as abundant and promising renewable energy sources (Ki et al., 2013). Biomass derived pyrolysis oil, namely bio-oils, possesses several environmental advantages over fossil fuels such as less undesirable gaseous emission of CO_2, SO_X, and NO_X (Isahak et al., 2012). In general, the bio-oil yield and properties can be significantly affected by pyrolysis temperature and the nature of feedstock (Chen et al., 2014). Although showing a great potential as an alternative energy source, the utilization of bio-oil has been limited due to some drawbacks such as high oxygen and water content, low heating value, and instability (Isahak et al., 2012). It is thus of importance to find effective solution for improving the biomass pyrolysis. The co-pyrolysis of biomass with other organic wastes seems to be a simple and effective way for such improvement not only in pyrolysis oil quality but also in yield (Kuppens et al., 2010; Abnisa and Daud, 2014). Biomass and organic wastes usually have different chemical and physical properties such as moisture, volatile matter,

* Corresponding author. UNBC/UBC Environmental Engineering Program, University of Northern British Columbia (UNBC), Prince George, British Columbia, V2N 4Z9, Canada.
E-mail address: Jianbing.Li@unbc.ca (J. Li).

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ash content, calorific value, porosity, and oxygen/hydrogen/carbon (O/H/C) molar ratio. The differing properties can change the reactivity and thermal characteristics of samples and products, and the formation of synergistic interaction during co-pyrolysis could then result in an improved pyrolysis product (Kar, 2011).

Many efforts have been made to investigate the co-pyrolysis of biomass with other wastes, such as waste tires, plastic wastes, and municipal sewage sludge (Samanya et al., 2012; Liu et al., 2013; Pinto et al., 2013; Martínez et al., 2014). For example, Brebu and Spiridon (2012) examined the co-pyrolysis of pinecone with synthetic polymers and found that the oil yield increased from 47.5 to 69.7 wt%. Önal et al. (2014) investigated oil production by co-pyrolysis of almond shell with high density polyethylene, and observed that the produced oil via co-pyrolysis had an improved heating value (i.e., by 38%) but a much-decreased oxygen content (i.e., by 86%) than those from biomass pyrolysis alone. Zuo et al. (2014) also observed that the higher heating value (HHV) of oil product was improved by the co-pyrolysis of sewage sludge with poplar sawdust as compared to pyrolysis of sewage sludge alone. However, so far there has been limited research investigating the co-pyrolysis of biomass with petroleum sludge. As a result, the present study aims to examine the effect of such co-pyrolysis. The wood waste (sawdust) was used as the biomass, and the refinery oily sludge waste was used as the petroleum sludge. The synergistic effect of co-pyrolysis on oil and char products was investigated. A number of factors such as sawdust percentage in the feedstock, pyrolysis temperature, and heating rate can affect the pyrolysis products (Abnisa and Daud, 2014). As a result, these three factors were examined in this study. The main and interaction effects of these factors on the yield of oil and solid char were evaluated through a response surface methodology (RSM) using a minimum number of experimental runs (Ahmadi et al., 2005). The quality of pyrolysis product was also analyzed. The results would provide valuable information for developing an effective strategy in terms of both resource recycling and oily sludge waste management.

2. Materials and methods

2.1. Materials

Wood waste (Douglas fir sawdust) was collected from a wood industry, and oily sludge was collected from tank bottom periodical cleaning at an oil refinery in western Canada. The sludge and sawdust samples were oven dried at 80 and 105 °C for 8 h to remove moisture, respectively (Tian et al., 2014). The sawdust was ground in a high-speed rotary cutting mill and then screened into particles in diameter of about 1 mm. Table 1 lists the sample properties and their test methods.

2.2. Experimental design

Three experimental factors were examined in this study, including sawdust percentage in the feedstock, pyrolysis temperature, and heating rate. By using Design Expert® 7.0, a five level-three variable central composite circumscribed (CCC) experimental design method was applied for arranging the co-pyrolysis experiments. The CCC had a factorial design and star points at a distance of ±1.682 from the central point. Table 2 lists the experimental levels (coded as ±1, 0, and ±1.682) and their real values. By using the CCC design method, a total of 17 experiments (8 factorial points, 3 center points, and 6 star points) were required. Each experiment arranged at the center of experimental domain (i.e., run # 15–17) was repeated for three times in order to estimate the pure error. The analysis of variance (ANOVA) was used for analyzing the experimental data. Multiple linear regression analysis was performed to fit a quadratic poly-nominal model:

$$Y = \beta_0 + \sum_{i=1}^{3}\beta_i x_i + \sum_{i=1}^{3}\beta_{ii}x_i^2 + \sum_{i=1}^{2}\sum_{j>i}^{3}\beta_{ij}x_i x_j \quad (1)$$

Where Y is the response variable (i.e. Y_o for pyrolysis oil and Y_c for char yield); x_i or x_j is the independent variable (i.e. experimental factor); β_0, β_i, β_{ii}, and β_{ij} are the intercept, linear, quadratic, and interaction coefficients of the model, respectively. In this study, the following symbols were used to represent the experimental factors: (A) the mass percentage of sawdust in feedstock (%), (B) pyrolysis temperature (°C), and (C) heating rate (°C/min). The pyrolysis oil or char yield was thus assessed as the sum of a constant, three first-order effects (terms in A, B, and C), three interaction effects (terms in AB, AC, and BC), and 3 s-order effects (A^2, B^2, and C^2). Only the terms found statistically significant were included in the model. The optimal co-pyrolysis condition for maximum oil yield was then identified using the numerical optimization function of Design Expert® 7.0 software.

2.3. Pyrolysis procedure

Each pyrolysis experiment was carried out in a fixed-bed tube furnace reactor (quartz tube length: 600 mm, Ø: 50 mm; MTI Corp.® GSL-1100X) under atmospheric pressure (Fig. 1). About 20 g of feedstock (i.e., sawdust and oily sludge mixture) was put into the

Table 1
Properties of sawdust and oily sludge.

Analysis	Value		Analytical methods
	Sawdust (raw)	Oily sludge (raw)	
Proximate analysis:			
Ash content (at 750 °C) (wt.%)	1.0	10.3	ASTM D3174
Moisture content (wt.%)	6.2	22.4	ASTM E871
Volatile matter (wt.%)	76.5	56.5	ASTM E872
Fixed carbon [a] (wt.%)	16.3	10.8	
Ultimate analysis:			
Carbon, C (wt.%)	47.0	60.5	ASTM D5373
Hydrogen, H (wt.%)	5.8	17.6	ASTM D5373
Oxygen [a], O (wt.%)	47.2	21.8	ASTM D5373
Nitrogen, N (wt.%)	0.01	0.05	ASTM D5373
Higher heating value (MJ/kg)	17.5	43.7	ASTM D5865/E711

[a] Calculated by mass difference.