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Fast co-pyrolysis of waste newspaper with high-density polyethylene for high yields of alcohols and hydrocarbons

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ABSTRACT

Waste newspaper (WP) was first co-pyrolyzed with high-density polyethylene (HDPE) using pyrolysis-gas chromatography/mass spectrometry (Py-GC/MS) to enhance the yields of alcohols and hydrocarbons. The effects of WP: HDPE feed ratio (100:0, 75:25, 50:50, 25:75, 0:100) and temperature (500–800 °C) on products distribution were investigated and the interaction mechanism during co-pyrolysis was also proposed. Maximum yields of alcohols and hydrocarbons reached 85.88% (feed ratio 50:50 wt.%, 600 °C). Hydrogen supplements and deoxidation by HDPE and subsequently fragments recombination result in the conversion of aldehydes and ketones into branched hydrocarbons. Radicals from WP degradation favor the secondary crack for HDPE products resulting in the formation of linear hydrocarbons with low carbon number. Hydrocarbons with activated radical site from HDPE degradation were interacted with hydroxyl from WP degradation promoting the formation of linear long chain alcohols. Moreover, co-pyrolysis significantly enhanced condensable oil qualities, which were close to commercial diesel No. 0.

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1. Introduction

Massive annual output of waste daily newspaper reached almost five million tons in China. Current recycling processes for this waste (e.g. farm incineration, burial or disposal in landfills) have caused serious environmental issues and prompted great interest on clean energy recycling method. In recent years, thermal conversion process such as pyrolysis applied to those wastes for bio-fuels production attracted great attention due to the increasing bio-energy demands and growing environmental pollution caused by the fossil fuels (Mohan et al., 2006; Yanik et al., 2007).

Almost all biomass can be converted into bio-fuels (e.g. bio-gas, bio-oil, or bio-char) via pyrolysis. Previous studies have showed that fast pyrolysis process (heating rate $>1000\text{ °C s}^{-1}$) has potential for converting the lignocellulosic biomass into carbon neutral fuel and intermediates (Pattiya, 2011; Wang et al., 2012). This process could produce bio-oil with high yield (65–75 wt.%) Liu et al., 2014 and high energy density which are four to seven times as compared to the overall energy density of the biomass alone (Kanaujia et al., 2014). However, fast pyrolysis of biomass alone produced highly oxygenated compounds in its derived bio-oil due to high oxygen

content in the feedstock, resulting in many unstable properties when it used as bio-fuel (Oasmaa et al., 2015). For example, carbonyl compounds in bio-oil cause instability and corrosion in engines; the components with high molecular weight in bio-oil result in high viscosity. Moreover, extremely low content of hydrocarbons and high water content in bio-oil lead to low heating value (Sundqvist et al., 2016). Those shortcomings greatly limited its application on fuel area used in standard engines.

Previous studies have demonstrated that bio-oil derived from biomass was upgraded into drop-in bio-fuel via catalytic cracking or hydrodeoxygenation (HDO) Liu et al., 2014; Wan and Wang, 2014. Both of the two upgrading processes lead to the removal of most oxygen (more than 90 wt.%) in the feedstock and favor the production of diesel and gasoline range fuels. However, the catalyst deactivation has greatly limited the application of those two upgrading process (Choudhary and Phillips, 2011). Moreover, the process of hydrodeoxygenation is highly hydrogen intensive and easy to form the compounds with high molecular weight and coke (Mercader et al., 2011).

In recent years, co-pyrolysis of biomass (e.g. lignocellulosic biomass and its components) with hydrogen-rich synthetic polymers was considered as an effective way to enhance the bio-oil yield and its fuel properties (Rutkowski, 2011; Brebu and Spiridon, 2011; Abnisa et al., 2013; Cornelissen et al., 2008). The content of carbon, hydrogen, and oxygen in the feedstock are re-adjusted resulting in

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positive synergistic effects on its derived products. However, the interaction mechanism during fast co-pyrolysis is largely unknown due to the presence of series of competing reactions. Moreover, the effect of fast co-pyrolysis on the yield of bio-fuel intermediates, especially alcohols and hydrocarbons, is also unclear. Previous studies have showed that cellulose gives higher liquid yield in comparison to lignin and lignocellulosic biomass since lignin mainly produced solid products during fast pyrolysis. Especially, Brebu et al. (2010), Brebu and Spiridon (2012) reported that co-pyrolysis of lignin with four synthetic polymers only gave the liquid product yields of 53.2–62.7 wt.% and also found that the liquid product yields from co-pyrolysis of pine cone with synthetic polymers reached 63.9–69.7 wt.%, while the highest liquid product yield of 79.5 wt.% was observed from co-pyrolysis of cellulose with polyethylene in Solak's study (Solak and Rutkowski, 2014a, 2014b). Moreover, our previous study has demonstrated that slow co-pyrolysis of waste newspaper (WP) with high-density polyethylene (HDPE) can enhance the yield of liquid product by 31.59% and significantly improve its fuel properties (Chen et al., 2016).

Therefore, waste newspaper which mainly consists of cellulose was selected in the present study in order to enhance the liquid product yield. HDPE was selected as hydrogen donor owing to its broad sources, low price, and high relative hydrogen content. Co-pyrolysis of WP with HDPE was carried out using pyrolysis-gas chromatography/mass spectrometry (Py-GC/MS), aiming at producing high yield of bio-fuel intermediates. The effects of feed ratio and temperature on co-pyrolysis derived products, especially alcohols and hydrocarbons, were evaluated. Such understanding is essential for effective utilization of wastes and development of its application in fuel area.

2. Material and methods

2.1. Materials

The feedstock used in this study contained WP and HDPE. WP was collected from local newsstand in Nanjing city. HDPE was supplied by Yangzi (Nanjing) Chemical Plastic CO., Ltd. Both of the two materials were first milled by a grinder then sieved into a particle size of less than 150 μm using a mesh screen. Then the treated WP and HDPE were mixed at the WP:HDPE mass feed ratio of 100:0, 75:25, 50:50, 25:75, 0:100 and rolled by a blender for 12 h to avoid the dispersion effect of newspaper, and then dried for 8 h at 80 $^{\circ}\text{C}$ in order to avoid softening of HDPE. For the analysis of different pyrolysis temperature, all mixed samples are obtained from the same rolled batch. Some typical properties of the feedstock were presented in Table 1.

Table 1
Ultimate and proximate analyses for the feedstock.

	WP	HDPE
<i>Elemental analysis,^a wt.%</i>		
C	39.78	85.43
H	5.50	14.21
O ^c	54.62	0.15
N	0.10	0.08
H/C molar ratio	1.66	1.99
O/C molar ratio	1.03	0.001
Calorific value, ^b kJ/g	14.57	38.66
<i>Proximate analysis, wt.%</i>		
Volatile	72.8	100
Moisture	6.5	–
Ash	11.2	–
Fixed carbon ^c	9.5	–

^a Dry basis and ash-free.

^b Dry basis.

^c Calculated by difference.

2.2. Fast pyrolysis procedure

Fast co-pyrolysis of WP with HDPE was performed in a pyrolyser, CDS pyroprobe[®] 5200 model. Approximately 0.5 mg of the sample was placed in the quartz tube for each experiment. Then the sample was subjected to fast pyrolysis at a heating rate of 20 $^{\circ}\text{C ms}^{-1}$ and the final temperature (500 $^{\circ}\text{C}$, 600 $^{\circ}\text{C}$, 700 $^{\circ}\text{C}$, 800 $^{\circ}\text{C}$) was hold for 50 s. The pyrolysis vapors released from quartz tube flowed into an Agilent 6890N gas chromatograph (GC) coupled with 5975 inert XL mass selective detector (MSD) instrument via the carrier gas of high-purity helium (99.999%) at a constant flow rate of 1.0 mL/min. A capillary column named HP5-MS (30 m \times 0.25 mm \times 0.25 μm , 5%-phenyl-95%-methyl polysiloxane) was used for GC separation. The GC/MS program is: ionization energy, 70 eV; injector temperature, 240 $^{\circ}\text{C}$; split ratio, 50:1; m/z range, 35–500 amu, volume of injected sample, 0.2 μL ; initial column temperature, 40 $^{\circ}\text{C}$, hold for 2 min; heating rate, 12 $^{\circ}\text{C}/\text{min}$; final temperature, 260 $^{\circ}\text{C}$, hold for 10 min. carrier gas, high-purity helium of 99.999%, 1 mL/min. Each experiment was repeated thrice and only their average values were used for discussion.

2.3. Products analysis

Most of the chromatographic peaks were identified based on the NIST library, and some were referred to the Wiley library and previous literatures (Brebu and Spiridon, 2011; Abnisa et al., 2013). Char yield was calculated using a high accuracy microbalance. Then quantitative analysis was performed for those compounds.

Theoretical value was defined via the additivity rule calculated from the products yield corresponding to the individual components as seen in Eq. (1) (Xu and Huang, 2010).

$$y = (x_1 \cdot w_1 + x_2 \cdot w_2) \quad (1)$$

where y represents the theoretical value; x represents the experimental products yield from individual WP and HDPE pyrolysis; w is the mass proportion of WP and HDPE in the feedstock.

Aliphatic hydrocarbons and aromatic hydrocarbons derived from fast co-pyrolysis were denoted as ALH and ARH, respectively.

2.4. Condensable oil analysis

A fluidized bed reactor was applied for fast co-pyrolysis of WP and HDPE. The reactor and collector diagram and its specification were referred to previous study (Kim et al., 2014). The reactor equipment was composed by a feeder, an injection auger, and a stainless steel reactor (height of 0.34 m, inner diameter of 38.1 mm). Silica sand (average diameter of 520 μm) was selected for heat carrier in the reactor and N_2 was used as the sweep gas. The feed ratio of the feedstock was 50 g/h and the estimated residence time of the released vapor was 1.2 s. The reactor was heated indirectly by electricity to a pyrolysis temperature of 600 $^{\circ}\text{C}$. A pipe which connects reactor and collector was preheated to 350 $^{\circ}\text{C}$ in order to avoid the condensation of pyrolysis gas during the experiments.

Oil derived from fast pyrolysis was recovered from electrostatic precipitator (organic phase with high molecular-weight compounds) and condenser (aqueous phase with low molecular-weight compounds) which are located downstream of the reactor. Oils in the present study referred to the sum of the condensable liquid products obtained from those two collectors.

Water content in aqueous phase was determined by a Karl-Fischer Titrator (KEM, MKS-500) with Hydranal-composite 5 K solution, while water content in organic phase was measured by a Mettler Toledo Thermogravimetry/Differential Scanning

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