



Preparation of liquid chemical feedstocks by co-pyrolysis of electronic waste and biomass without formation of polybrominated dibenzo-p-dioxins

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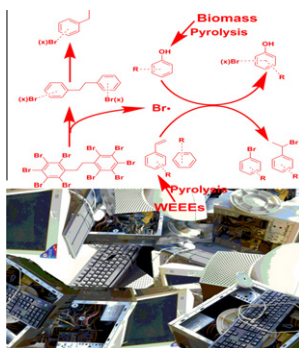
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HIGHLIGHTS

- ▶ Proposed a novel method to dispose the WEEEs and recover chemical feedstocks.
- ▶ Improved the conversion efficiency of WEEEs by adding a give portion of biomass.
- ▶ Interpreted the conversion mechanism of co-pyrolysis of WEEEs and biomass.
- ▶ Proposed a strategy to block the formation pathway of PBDD/Fs from BFRs.

GRAPHICAL ABSTRACT

Waste electrical and electronic equipments (WEEEs) have become one of the fastest growing waste streams around the world. An environment benign co-pyrolysis method of WEEEs and biomass to reclaim renewable chemical feedstocks was proposed. The synergistic effect of the WEEEs and biomass improves the conversion efficiency of WEEEs and block-ups the formation of PBDD/Fs, and the mechanism of co-pyrolysis was discussed.



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ABSTRACT

The co-pyrolysis of waste electrical and electronic equipments (WEEEs) and waste biomass to obtain pyrolysis-oil, a liquid fuel or chemical feedstock, was carried out in the present work. The pyrolysis-oil yield of co-pyrolysis reached 62.3% which was significantly higher than those of pyrolysis of WEEEs and biomass alone (i.e., 53.1% for WEEEs and 46.3% for biomass), suggesting that synergistic effects of the WEEEs and biomass happened during the co-pyrolysis process. The pyrolysis-oil mainly contained aromatic compounds, including many aromatic hydrocarbons. More than 90 wt.% of bromides were enriched in pyrolysis-oil and char, which is easily to be recovered by further treatments, and no polybrominated dibenzo-p-dioxins and furans (PBDD/Fs) were detected in all products which may be attributed to the blocking of PBDD/Fs generation under special reductive environment of pyrolysis. This work provided a green and environmentally friendly approach for the disposal of the WEEEs as well as resource recovery.

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1. Introduction

Along with the continuous expansion of electronic and electrical equipments industrial market, waste electrical and electronic

equipments (WEEEs) have become one of the fastest growing waste streams around the world. As the United Nations Environment Program estimates, the world generates 20–50 million tons of WEEEs each year, and their amounts are rising three times faster than those of other municipal waste (Huang et al., 2009). Distinct from other forms of municipal or industrial waste, the WEEEs contain both valuable and hazardous materials which need recycling

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and special handling methods to avoid environmental pollution and detrimental effects on human health (Söderström and Marklund, 2002). However, due to lack of facilities, high labor costs and tough environmental regulations, it is not economically friendly to recycle materials from WEEEs. Instead, most of the WEEEs are currently landfilled or incinerated directly in many developing countries (Alston and Arnold, 2011), which will bring about serious environmental pollution. For example, the WEEEs landfilling requires large space, and the resulting leachate usually contains many pollutants which may contaminate the surrounding environment and groundwater (Spalvins et al., 2008). On the other hand, incineration of WEEEs will bring about serious air pollution due to the emission of particulate matter (e.g., PM 2.5) and dioxin-like compounds during the combustion process. More seriously, the brominated flame retardants (BFRs), which are widely used in the electronic and electrical equipments to reduce the possibility of fire under the thermal stress, have great potential to form highly toxic POPs under incineration conditions, such as polybrominated dibenzo-p-dioxins and furans (PBDD/Fs).

Fast pyrolysis, as an economically feasible and environmentally friendly technology for treatment of WEEEs and other organic waste, has more advantages than landfilling and incineration, as the energy consumption of the fast pyrolysis is very low (only about 10% of energy content of the WEEEs was consumed in the fast pyrolysis process). Furthermore, the emission of harmful gases is much lower than that of the incineration process (Bhaskar et al., 2002). The conversion of the WEEEs into fuel or chemical feedstocks by the fast pyrolysis is a promising method for preserving valuable resources as well as protecting the environment by limiting the amount of non-degradable waste. During the pyrolysis, the WEEEs are thermal-decomposed and converted into oil, gas, and char products at a moderate temperature (usually 700–900 K) with high heating rate (more than 300 K s^{-1}) in an oxygen free system. The produced oil can be used as chemical feedstocks or as fuels (Hall et al., 2007; Vispute and Huber, 2009). Alston et al. (2011) and Hall and Williams (2006a,b) have demonstrated that fast pyrolysis can be successfully used to process WEEEs, and the amount of produced oil is usually variable with the composition of the WEEEs and experimental conditions.

However, the yield of oil in the fast pyrolysis of the WEEEs is often constrained by their compositions of low hydrogen to carbon molar ratio. To increase the oil yield, it is necessary to provide additional hydrogen source to WEEEs pyrolysis process from other feedstocks. The lignocellulosic biomass, which has high hydrogen to carbon molar ratio, can act hydrogen-donors in the co-pyrolysis process of biomass and WEEEs (Zhang et al., 2007). Furthermore, the synergy effects between the biomass and WEEEs which promotes the formation of volatile matters is potentially existed, and causal to better thermal conversion performance of the WEEEs. Therefore, it is desirable to co-pyrolysis the biomass and WEEEs to achieve a high thermal conversion efficiency, to maintain a green utilization level of WEEEs and to minimize the adverse impact of WEEEs reclamation on environment.

Besides the resource recovery, the transformation the pollutants (e.g., BFRs) in the fast pyrolysis also need to investigate. The BFRs are persistent and bioaccumulative organic pollutants, and their fate in the combustion process has been well investigated (Gullett et al., 2009; Sakai et al., 2001). However, the fate of BFRs in the co-pyrolysis process is unclear and needs in-depth study. Unlike combustion process, the fast pyrolysis process is proceeding in an oxygen-free atmosphere, and the BFRs may experience a very different process compared to that in combustion. The migration and transformation behaviors of the BFRs may provide some basic information for avoidance of PBDD/Fs occurrence and selectively changing the contents of the bromine in pyrolytic products so as

to conveniently recover it. To our best knowledge, such information has not been reported in the literature till now.

In this work, the WEEEs were co-pyrolyzed with fir sawdust, an abundantly available waste biomass, to produce pyrolysis-oil. The main objective of this work is to produce the pyrolysis-oil by co-pyrolysis of biomass and WEEEs and investigate the fate of BFRs during the pyrolysis process. To this end, (1) the effects of temperature and the ratio of WEEEs and sawdust on the yield of pyrolysis-oil were investigated; (2) the total pyrolysis products (i.e., pyrolysis-oil, char, and gas) were analyzed by ICP-MS, GC-MS and XPS; (3) the bromine-containing compounds were characterized and a mechanism for the transformation of BFRs in the pyrolysis process was proposed; and (4) the blocking-up of PBDD/Fs generation was discussed. This study may provide some basic insight on the emission reduction of Br-containing POPs during environmentally friendly utilization of WEEEs.

2. Methods

2.1. Materials

The WEEEs samples were collected from a local waste recycling plant. Before use, the plastic fraction of WEEEs was crushed by a high-speed rotary cutting mill and the particles with the size smaller than 0.12 mm (120 mesh) were collected for the further experiments. The BFR added in the WEEEs is 2,2',3,3',4,4',5,5',6,6'-decabromobiphenyl (DBDPE), an alternative to the polybrominated diphenyl ethers (PBDEs) because of its high stability and excellent flame retarding effect (Wu et al., 2012). The general characteristics of DBDPE are shown in Table S1 of supporting information. The biomass sample (fir sawdust) was obtained from a local timber treatment plant. The sawdust was crushed and screened to limit the particle size smaller than 0.12 mm for the further use. The proximate analysis of the biomass and WEEEs was referred to ASTM-D5142, and their main element compositions (C, H, N, and O) were analyzed using an elemental analyzer (VARIO EL III, Elementar Inc., Germany). The mineral elements in the biomass and WEEEs were determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES, Optima 7300 DV, Perkin Elmer Co., USA). The thermal stability of the WEEEs and biomass was characterized with a DTG-60H/DSC-60 thermogravimetric analyzer (Shimadzu Co., Japan).

2.2. Co-pyrolysis of WEEEs and biomass

Before pyrolysis, the sawdust and WEEEs was mixed uniformly at different ratios as feedstock. The fast pyrolysis experiments were carried out in a vertical drop fixed-bed reactor described in our previous work (Liu et al., 2011). A feedstock of 5.0 g was firstly placed in the feed pipe in each experiment, and then a nitrogen gas flow of 400 mL min^{-1} was provided and maintained for 20 min to remove air from the pyrolysis system. Soon after the pyrolysis temperature reached the setting value (e.g., 723–873 K), the feedstock was fed into the quartz reactor via a piston. The temperature of the feedstock increased from room temperature to the reactor temperature in its falling process (about 1–2 s) at a heating rate over 300 K s^{-1} . The volatiles produced in the pyrolysis process were swept out by nitrogen gas at a flow rate of 200 mL min^{-1} , and condensed by a cold ethanol-ethylene glycol mixture (1:1, v/v) to produce pyrolysis-oil. The incondensable components in the volatiles were introduced into two absorption bottles filled with acetone and NaOH solution (1.0 mol L^{-1}) in row. When the pyrolysis process was completed, the reactor was immediately moved out from the heating zone, and cooled to room temperature in a nitrogen gas

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