



Fast pyrolysis of biomass and waste plastic in a fluidized bed reactor



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HIGHLIGHTS

- A mixture of red oak and HDPE is fast pyrolyzed in a fluidized bed.
- Optimum pyrolysis temperature was 625 °C.
- A significant synergistic effect between red oak and HDPE was found.
- Co-pyrolysis reduces char formation from red oak.
- Pyrolysis char had a lower BET surface area than red oak biochar.

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ABSTRACT

Co-pyrolysis of red oak and high density polyethylene (HDPE) was conducted in a laboratory-scale, continuous fluidized bed reactor in a temperature range from 525 to 675 °C. Pyrolysis products, including two fractions of pyrolysis-oil, non-condensable gases and char were analyzed to assess the influence of pyrolysis temperature and co-feeding of biomass with HDPE. It was found that increasing pyrolysis temperature up to 625 °C promoted the production of pyrolysis-oil and its yield reached 57.6 wt%. Further increase in pyrolysis temperature caused the cracking of pyrolysis-oil to form light gases rich in hydrocarbons. Organic phase of pyrolysis-oil produced from plastic-biomass mixture (PBM) had a higher heating value (HHV) up to 36.6 MJ/kg contributed by the additive effect of HDPE-derived aliphatic hydrocarbons. A significant synergetic effect was also observed during co-pyrolysis. Co-pyrolysis with HDPE increased the production of furan, acids and water from red oak. Co-presence of HDPE also inhibited char formation from red oak and improved the HHV of the resulting char. The char produced from co-pyrolysis had a significantly lower BET surface area than red oak biochar. Not only did HDPE-derived particulate matter blocks the pores, the synergetic interaction also resulted in the formation of large and shallow micro-pores on the char surface.

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1. Introduction

Each year the US alone produces 250 million tons of municipal solid waste (MSW). Among it, over 50% of the non-recyclable MSW ends up in landfill sites [1]. The landfilled MSW takes away valuable land and creates numerous potential environmental problems. In fact, the discarded MSW represents a tremendous energy source. Waste-to-energy (WTE) technologies can mitigate negative impacts of MSW and provide sustainable energy from low-cost feedstock. Examples of these technologies include incineration, gasification, anaerobic digestion, and pyrolysis [2,3]. Pyrolysis depolymerizes dry feedstock under an

oxygen free environment. When the pyrolysis temperature is moderately high (450–550 °C) [4], the volatiles arise from pyrolysis process can be condensed to become liquid product, called pyrolysis-oil [5]. Unlike other technologies that produce heat or gases, pyrolysis-oil is transportable liquid and can be upgraded to transportation fuels or other platform chemicals [6]. Another advantage of the pyrolysis process is that it has low requirements for the feedstock type and reactor design, thus technology is relatively easy to scale up.

While MSW consists of many different types of materials, biomass and plastics make up a majority of the composition [1]. When biomass is pyrolyzed alone, it produces a number of oxygenated products, such as sugars, aldehydes, ketones, acids and phenols. The presence of oxygen in the pyrolysis-oil (resulting from an abundance in the biomass feedstock) lowers the heating value and also causes thermal instability and corrosiveness [7].

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On the other hand, plastic wastes are rich in hydrogen and contain much less oxygen than biomass. High density polyethylene (HDPE), the most commonly used plastic for example, has virtually no oxygen. Thus, compared to pyrolyzing biomass alone, co-pyrolyzing biomass and waste plastics increases carbon and hydrogen contents in the feedstock and could be beneficial in improving the quality of pyrolysis-oil. As a result, higher quality pyrolysis-oil could potentially reduce the costs associated with catalytic hydrodeoxygenation, which is required to process it into hydrocarbon fuels [8].

Co-pyrolysis of biomass with different plastics has been investigated extensively [8–22]. Biomass and the plastics were often placed inside of batch reactors or fixed bed reactors and slowly pyrolyzed for an extended period of time in a discontinuous mode [8]. For example, Costa et al. [9] co-pyrolyzed rice husk and polyethylene in a batch reactor at 350–430 °C for up to 60 min and reported that the thermal conversions of biomass and PE are facilitated by the presence of each other. Martinez et al. [12] also slowly pyrolyzed biomass and synthetic polymers and found that the viscosity and acidity of pyrolysis-oil decreased whereas the heating value increased compared to that of pyrolysis-oil obtained when pyrolyzing biomass alone. It was also reported that the yield of pyrolysis-oil was much higher than the theoretical sum of pyrolysis oils produced from biomass and plastics when they are independently pyrolyzed [13]. Recently, Sajdak et al. [19–21] thoroughly investigated co-pyrolysis of biomass and polypropylene and concluded that co-pyrolysis has a significant synergistic effect. In their study, the mixed feedstock was pyrolyzed in a batch reactor for 50 min at a heating rate of 5 °C/min.

It is noteworthy that in general, maximum yield of pyrolysis-oil is achieved from biomass upon fast pyrolysis [23] since slow pyrolysis of biomass usually promotes the formation of char and light gases instead of pyrolysis-oil [24]. During fast pyrolysis, the feedstock is rapidly heated (>100 °C/s) and pyrolysis vapor is instantly swept away from the reactor zone and quenched. The vapor retention time is usually less than 2 s in fast pyrolysis to limit secondary reactions that decreasing the amount of the condensable vapor. Although fast pyrolysis of biomass alone was extensively studied, very few investigated fast co-pyrolysis of biomass and plastics [10,15,16,18,22,24] in continuous mode. It was reported that the synergetic effect among biomass and plastics is negligible during fast pyrolysis due to the short reaction time in the reactor [10,22]. A contradictory result, however, is reported by Martinez et al. at a study using an auger reactor [12].

It should also be noted that fast co-pyrolysis of biomass and plastics was mostly conducted at the optimum temperature for fast pyrolyzing biomass (450–500 °C). However, the optimum pyrolysis temperature of biomass is often too low for completely decomposing plastics during fast pyrolysis since some plastics, such as HDPE, degrade at much higher temperatures than biomass does [25].

In this study, red oak and HDPE pellets were co-pyrolyzed in a lab-scale, continuous fluidized bed reactor. The estimated heating rate in the fluidized bed is 600 °C/s, which is typical for fast pyrolysis. Fast pyrolysis at below 500 °C in the reactor is not sufficient to completely depolymerize HDPE to volatiles due to the short reaction time. As a result, the melted plastic either forms agglomerates with the fluidizing sand or biomass thus developing defluidization inside of the fluidized reactor. Thus it was determined that the fast co-pyrolysis of biomass and HDPE has to be conducted in a temperature range higher than the optimal temperature of biomass pyrolysis. In this work, pyrolysis products, including two fractions of pyrolysis-oil, non-condensable gases and pyrolysis char were analyzed using comprehensive analytical methods and the results were compared with the products produced from pyrolysis of red oak alone.

2. Material and methods

2.1. Feedstock

High density polyethylene (HDPE) pellets were obtained from USI Corporation, Taipei. The pellets, 4 mm in diameter and 2 mm in thickness, are made from recycled plastics. Northern red oak (*Quercus Rubra*) was purchased from Wood Residues Solutions (Montello, WI). The bark free chips were first ground by a mill cut and then sieved to a constant size range between 250 and 400 µm. Proximate and ultimate analyses of the feedstock are given in Table S1.

2.2. Pyrolysis

Co-pyrolysis of red oak and HDPE was conducted using a laboratory-scale continuous fluidized bed reactor. The schematic diagram of the reactor system and its specification can be found elsewhere [26]. Specifically, the reactor system consists of a feeder, an injection auger, and a stainless steel reactor that is 0.34 m in height and 38.1 mm in inner diameter. Silica sand from 410 to 600 µm was used as the heat carrier in the reactor and nitrogen gas was used as the sweep gas. The feed rate of material was 60 g/h and the estimated vapor residence time in the reactor was 1.1 s [27].

The separation and collection of pyrolysis products were conducted by two cyclones, an electrostatic precipitator (ESP) and condensers that are located downstream of the reactor. Char and ash particles were removed by two cyclones connected in series. Liquid nitrogen was sprayed to the pyrolysis vapor prior to the vapor stream entering the ESP to reduce the temperature of the vapor steam to 90 °C. The aerosols of relatively high molecular-weight (MW) compounds in the vapor were collected at a collection bottle attached to the end of the ESP. This fraction of pyrolysis-oil is referred as organic phase in this study. The light MW compounds in the pyrolysis vapor were condensed and recovered at further downstream using a condenser chilled to –10 °C. This light MW fraction of pyrolysis-oil is referred as aqueous phase.

Pyrolysis char was collected in the cyclones. The yield of char was determined by weighing the sand bed and two cyclones before and after each experiment. The yield of pyrolysis-oil was determined by measuring the weight difference of the pyrolysis-oil collection system including pipes, vessels and containers. The composition of non-condensable gases (NCGs) in the exhaust stream was measured with a micro-Gas Chromatogram (GC) (Varian CP-4900) calibrated for nitrogen (N₂), hydrogen (H₂), carbon oxides (CO₂, CO), and hydrocarbon gases up to C₃ which include ethylene (C₂H₄), ethane (C₂H₆), and propane (C₃H₈). A drum-type gas meter (Ritter, Germany) and the ideal gas law were used to determine the volume of NCG. Since the hydrocarbon gases with C₄₊ were not calibrated in this study, the total yield of NCGs was reported by subtracting the yields of pyrolysis-oil and char from 100%.

According to the EPA report, the ratio of plastics to biomass is 1:4 in general MSW [23]. Thus, a mixture of the feedstock consisting of 20% HDPE and 80% red oak was prepared and pyrolyzed at 525, 575, 625 and 675 °C, respectively. Red oak alone was also pyrolyzed at 575 °C for the comparison. All the pyrolysis tests were duplicated and average mass yields were reported in this study. The standard errors between two runs at the same conditions were all below 5%.

2.3. Characterization of pyrolysis products

The pyrolysis-oil and char were characterized using the analytical methods described next. The CHNS elemental analysis of

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