



Fast co-pyrolysis of low density polyethylene and biomass residue for oil production



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ABSTRACT

Co-pyrolysis of low density polyethylene (LDPE) with three kinds of biomass was performed in a drop-down tube reactor. The synergistic effect was investigated by comparing the experimental relative yield and characteristics of oil with the calculated ones based on the results from the pyrolysis of the individual components in the mixture. It is found that the maximum oil relative yield in the case of co-pyrolysis process was obtained at 600 °C, which was significantly higher than the optimum temperature of biomass or LDPE pyrolyzed alone. Although the inorganic elements in biomass improved the decomposition of LDPE, more organics with larger molecule weight (>C12) were produced due to the further reaction between the decomposition products from LDPE and biomass. The synergistic effect was positive for the production of aliphatic compounds. However, it could be positive or negative for the production of aromatic compounds, which depended on the type of biomass in the feedstock. The significant removal of aldehydes, acids, ethers, furans, ketones, phenols and sugars from the final co-pyrolysis oil, in which there was a significant increase of alcohols, was achieved.

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1. Introduction

Biomass residue is considered as an environmental friendly energy since it is renewable and CO₂ neutral. The production of bio-oil is one of the most remarkable topics in the utilization of biomass residue. However, due to the high oxygen content in biomass, the bio-oil has low calorific value with corrosion problems and instability [1]. On the other hand, among the municipal solid wastes, the plastic waste is a recyclable resource with considerable amount. If the plastic waste ends up in landfill, it will be just as other non-recyclable municipal solid wastes, and create numerous potential environmental problems. Co-utilization of plastics and biomass residue by thermochemical conversion may overcome such problems and provide sustainable energy.

Pyrolysis is considered as clean thermochemical conversion technologies to provide energy and chemicals from biomass residue, and gained more and more attention [2–5]. In recent years,

a lot of researches have focused on the co-pyrolysis of plastics and biomass since a positive effect was always observed when the mixture of plastics and biomass was used as feedstock [6–28]. It has been found that the increased liquid yield from co-pyrolysis without any catalyst compared to the theoretical one according to the yields from the individual pyrolysis of biomass and plastics [11–16]. Furthermore, because of the addition of plastics as hydrogen content resource, the quality of oil obtained from co-pyrolysis was higher than that from the pyrolysis of biomass alone [17,18]. It is observed that an increase of hydrocarbon content and a decrease of oxygen content in the co-pyrolysis oil, resulting in an increased calorific value [18,19]. Generally, the oxygenated organics reduced the quality of pyrolysis-oil were alcohols, aldehydes, phenols, ketones, esters, sugars and so on [3]. It is reported that the content of phenolic compounds was significantly reduced [19,20], while the yields of furan and acids were promoted by the co-pyrolysis of woody biomass with polystyrene [21]. However, it is not so clear that the effect of the addition of plastics on the removal of other oxygenated organics generally existed in the bio-oil derived from the pyrolysis of woody biomass.

In the researches on co-pyrolysis mentioned above, most experiments were performed at a slow heating rate (<100 °C/s), which is

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called slow pyrolysis. Compared to the slow pyrolysis, fast pyrolysis with a heating rate of several hundreds °C/s generally promoted the yield of bio-oil instead of char and gas [22,23]. However, a contradictory result was reported on the synergistic effect during fast co-pyrolysis of plastics and biomass. Cornelissen et al. [26–28] and Xue et al. [21] reported an increased yield and calorific value of oil can be obtained from the fast co-pyrolysis of woody biomass and plastics, indicating a pronounced synergistic effect. Instead, Matsuzawa et al. [24] and Bhattacharya et al. [25] reported that the synergistic effect can be ignored during the fast co-pyrolysis due to the ultra-short reaction time.

In the present study, three kinds of biomass samples were separately co-pyrolyzed with a model chemical of plastic waste, i.e., low density polyethylene (LDPE), in a fixed bed reactor in order to investigate the effect of pyrolysis temperature on the oil yield and the synergistic effect on the oil compounds, especially the removal of oxygenated organics, during the fast co-pyrolysis, which were not obtained enough attention previously. It is expected to find valuable information on the optimum co-pyrolysis temperature and understanding the synergistic effect in the co-pyrolysis process in more details.

2. Experimental

2.1. Feedstock preparation

Three kinds of biomass, i.e. cedar wood, sunflower stalk and Fallopia Japonica stem (FJS), from Aomori prefecture Japan were mixed with low density polyethylene (Wako, Japan). The proximate and ultimate analyses of the biomass samples were presented in Table 1. One can see that many volatile matters are included in these biomass and the calcium and/or potassium are the main ash compositions, which have been found to have catalytic activity to accelerate the pyrolysis rate of biomass [29]. Moreover, all of them are easily obtained in Aomori prefecture Japan. Thus, they are suitable for production of bio-oil by thermal pyrolysis. Plastic wastes such as plastic bags, which are mainly made of LDPE, is one of the main daily life refuse in Japan [30]. In order to reduce the landfill use and recovery energy, a suitable thermal treatment technology is necessary to recycle LDPE waste.

In order to remove the moisture in the feedstock, all of biomass samples were dried for 24 h at 110 °C while LDPE powder was dried for 24 h at 80 °C at first. Three kinds of dried biomass samples were then separately mixed with dried LDPE powder in a ball mill

at 1:1 weight ratio to form an evenly distributed powder. The powder was pressed to tablet and then the tablet was gently crushed and sieved to the sample with particle size of 1–2.8 mm. The remaining moisture was analyzed by using MX50 moisture content analyzer (AND, Japan).

2.2. Pyrolysis setup

A dropdown tube pyrolyzer, which can conveniently achieve a rapid heating rate and short gas resident time, was used. The reaction system was precisely described in our previous work [5]. At first, 5 g of fine silica sand was filled into the reactor as bed material. Then, 0.5 g of the sample was put in the feeder section and flushed by argon to remove the air. Meanwhile, the reaction zone was heated to the desired pyrolysis temperature which was varied from 500 to 600 °C and kept stable for 1 h while the air was removed by argon gas to form an inert atmosphere inside the reaction zone. Pyrolysis process was started when the feeder valve was opened and let the sample fell down to the reaction zone. The sample can be heated to the pyrolysis temperature of 500–600 °C instantly. Pyrolysis process was held for 10 min. The pyrolysis oil including water was flowed out from the reactor by using 100 cm³/min of argon and condensed in two cold traps. The estimated gas resident time in the reaction zone was about 30 s. Non-condensable gases were then passed through a gas purifier and collected in a gas bag for further analysis using a gas chromatograph (TCD-GC, Agilent 7890A, U.S.) to determine the amounts of gases of H₂, CO, CO₂ and CH₄ [35]. The oil collected in the condensing section was analyzed using a gas chromatograph/mass spectroscopy (GC/MS QP2010, Shimadzu, Japan). Here, the products with boiling points lower than 300 °C can be detected. The water content of the obtained liquid was analyzed by using Karl-Fisher Titration method (MKS-500, KEM, Japan). Since some liquid products were found to be condensed in the pipeline and some possible hydrocarbon gases cannot be detected by GC. Here, the relative yield of oil product was calculated from mass balance after subtracted by the water content and the relative yields of detected gas and solid product. The percentage of each subgroup, for example, aliphatics, aromatics, phenols and so on, in the oil product can be detected by GC/MS, which was used for evaluating the results. The relative yield of each subgroup was obtained by the multiplying the peak area percentage in the GC/MS spectrum by the relative yield of oil product. Remaining solid in the reactor was calcined at 650 °C for 3 h and measured its weight before and after the calcination in order to quantify the amount of char and coke. The acetone-insoluble chemicals left in the reactor and condensers were named as the waxes as presented in Table 2. Similar waxes were also reported by Williams and Williams [31]. The waxes were immersed in 20 ml acetone for 24 h in order to extract the possible oil inside, and then separated from the acetone by centrifuge. After drying at 70 °C for 3 h, the weight of waxes was recorded.

Table 1

Proximate, ultimate and ash content analysis of the biomass samples.

Sample	Cedar	Sunflower	FJS
Ash	0.6	11.67	2.39
Volatile matter	86.9	87.07	88.2
Fixed carbon ^a	12.5	1.26	1.33
C	48.8	39.57	44.79
H	6.6	5.39	5.88
O ^a	43.0	53.61	49.03
N	1.4	1.18	0.28
S	n.d	0.25	0.02
CaO	50.68	16.30	50.24
K ₂ O	4.01	71.69	44.03
P ₂ O ₅	6.45	9.01	1.68
SO ₃	10.47	2.81	2.69
Fe ₂ O ₃	0.8	n.d	0.62
SiO ₂	6.83	n.d	0.70
SrO	0.04	0.1	0.04
Others	20.72	0.09	0

n.d: not detected.

^a Calculated by difference.

Table 2

Product distribution from the pyrolysis of different feedstocks at 600 °C (wt%).

Feedstock	Gas	Water	Tar/oil ^a	Char/coke	Waxes
LDPE	2.28	9.38	83.33	0.37	4.44
Cedar	14.70	25.74	38.83	20.73	–
Cedar/LDPE	7.98	15.94	64.08	9.32	2.69
Sunflower	16.17	27.62	29.94	31.31	–
Sunflower/LDPE	7.45	17.15	57.17	13.15	4.23
FJS	15.09	27.75	30.43	26.74	–
FJS/LDPE	8.02	16.69	58.96	13.47	2.67

^a Calculated by difference.

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