



## Investigation on the co-pyrolysis of waste rubber/plastics blended with a stalk additive



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### ABSTRACT

This study presents an experimental investigation of co-pyrolysis of discarded rubber and plastics in a tube furnace. The properties of the pyrolysis oil and solid residue under various blending ratios of rubber and plastics were investigated. The impacts of the stalk additive and the heating rate on the pyrolysis oil yield and feedstock-to-oil energy conversion efficiency (FOECE) were studied. Compared with the pyrolysis of rubber or plastics separately, the co-pyrolysis of rubber and plastics produced a higher oil yield with a higher oil heating value. When the mass fraction of the rubber was 60 wt%, the pyrolysis oil yield and FOECE reached their maximum values. When adding 10% stalk additive at a rubber-to-plastics ratio of 4:1, a higher oil yield was achieved, and the oil yield increased by 10.3 wt%, where the heating value of the pyrolysis oil reached 39.93 MJ kg<sup>-1</sup>. The stalk additive only had a slight impact on the solid residue and the completeness of the pyrolysis reactions. The stalk additive increased the FOECE and postponed the highest yield point to the 80 wt% rubber blend because the greater volatility promoted the pyrolysis reaction and the production of condensable gaseous components. A higher heating rate improved the oil yield. According to the elemental analysis, a small fraction of sulfur existed in the oil. The stalk additive increased the H/C ratio in the oil. Through the GC-MS analysis, the primary components in the pyrolysis oil from the rubber and plastics blend were identified as alicyclic hydrocarbons and aromatics, whereas the oxygen-rich stalk additive increased the contents of alcohols, esters, and ketones. Overall, the stalk additive significantly improved the yield and the FOECE when co-pyrolysis of rubber/plastic blends was performed.

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### 1. Introduction

The reclamation treatment of garbage, including municipal solid waste and industrial refuse, has become an important problem to solve the global energy crisis and a topic of environmental conservation studies. Garbage resource treatment technologies, such as biogas recovery from municipal solid waste [1,2], municipal solid waste composts [3,4], garbage incineration for power generation [5,6], and fuel or chemical material extraction by garbage gasification or liquefaction [7–9], have been widely developed and even commercially applied. However, most garbage is a mixture, which adds to the complexity of garbage treatment. Waste sorting is necessary, e.g., only degradable garbage can be used in biogas

recovery, and non-degradable waste should be handled using other methods. Waste incineration and pyrolysis belong to thermochemical treatment technology with advantages such as a high energy recovery rate, high attenuation ratio, etc. [5–9]. Compared with waste incineration, it is advantageous for non-degradable waste to be pyrolyzed to extract pyrolysis oil. The work temperature is lower and produces less dioxin and other pollutants. This technology can be applied to both degradable and non-degradable waste. Pyrolysis oil can be used as fuel and can also be used to extract chemical products with higher economic value [10]. In this paper, non-degradable waste mixtures (primarily discarded rubber and plastics) were treated with pyrolysis technology. Regarding the aspect of the reclamation treatment of junked tires, Martinez et al. [11] studied the co-pyrolysis of biomass with waste tires in a fixed bed and a rotary furnace. The free radical products in the co-pyrolysis interacted with each other and helped to form bio-oil with stable performance. Acevedo et al. [12] conducted

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**Table 1**  
Analysis of feedstock.

Properties	Rubber	Plastic	Cornstalk
Proximate analysis <sup>a</sup>			
Moisture (%)	2.34	0.66	3.52
Volatile (%)	61.23	63.48	75.75
Ash (%)	8.27	31.69	6.61
Fixed Carbon (%)	28.16	4.17	14.12
Element analysis <sup>a</sup>			
Carbon (%)	76.88	56.39	45.44
Hydrogen (%)	7.57	6.71	5.29
Oxygen (%)	2.39	1.75	38.27
Nitrogen (%)	0.72	0.50	0.62
Sulfur (%)	1.83	2.30	0.25
Heating value, LHV <sup>b</sup> , $Q_{\text{net}}/\text{MJ kg}^{-1}$	36.63	20.16	18.95

Note: LHV—Low heat value.

<sup>a</sup> As received basis.

<sup>b</sup> Dry basis.

co-pyrolysis of waste tires and coal in a fixed bed and obtained pyrolysis oil, whose primary contents were aliphatic and aromatic compounds. Tire crumb produced more limonene and BTX (benzene, toluene, and xylene). Jakab et al. [13] studied the co-pyrolysis of polypropylene plastics and additive (such as wood powder, lignin, cellulose, and charcoal) and noted that in the presence of these additives, particularly wood-derived materials, the reaction temperature caused by the low-temperature pyrolysis of plastics decreased, and wood-derived materials promoted the production of monomers and dimers. Regarding plastics garbage disposal studies, Syamsiro et al. [14] studied the pyrolysis of three types of discarded plastics and found that the types of raw materials greatly influence the yield rate and the quality of both liquid and solid products. HDPE (high-density polyethylene) waste produced the greatest liquid yield, and plastics produced oil with a higher heating value than that of biomass and inferior coal. Kumar et al. [15] used HDPE waste to perform catalytic cracking at the optimal condition, and pyrolysis oil with HC (C10–C25) was obtained, whose heating value reached 40.17 MJ/kg. Therefore, the oil produced by co-pyrolysis of discarded rubber and plastics has been recognized to be more feasible, and the approaches to optimize pyrolysis performance by biomass additives have also been studied. A small proportion of biomass additive can be added to the discarded rubber/plastics to optimize the pyrolysis technique of non-degradable waste further, improve the yield of pyrolysis oil extraction, and provide more economical and practical technical methods for non-degradable garbage treatment.

## 2. Materials and methods

### 2.1. Feedstock

The rubbers from junked automotive tires and plastics from collected packing bags were selected as feedstock. Cornstalk was used as an additive to improve yield. The materials were all crushed into powder with an average particle size of 0.30 mm.

Table 1 shows the results of the proximate analysis, element analysis, and heating value of the feedstock. The analysis was performed according to the Coal Analysis Standards (GB/T212-2008; GB/T213-2008; GB/T476-2001).

The rubber and plastics were blended at varying ratios, and the mass fraction of the rubber powder in the samples were 0 wt%, 20 wt%, 40 wt%, 60 wt%, 80 wt%, and 100 wt%. The mass of each sample was approximately 50 g. When cornstalk was added at a mass fraction of 10 wt%, the ratios of rubber to plastics were kept the same, and the total mass of the sample fuel was also 50 g, where the rubber/plastics mass was 45 g and the stalk mass was 5 g.

### 2.2. Experimental setup

Fig. 1 shows the schematic of the experimental facility. Pyrolysis was performed in a tube resistance furnace system, which primarily consists of a carrier gas supply, gas flow meter, heating and temperature control system, and condensation and collecting devices, which include two condensation pipes and two flasks. Ice water circulates in the interlayer of the condensation pipes to cool the flowing pyrolysis gas and collect the pyrolysis oil condensed from the gases. High-purity nitrogen was chosen as the carrier gas, and at the system end, a gas sampling bag was used for non-condensable gas collection.

### 2.3. Experimental procedure

Experiments were conducted using the following procedure. Firstly, the sample was set in the tube furnace;  $\text{N}_2$  was used to purge the pipes for 15 min at a flow rate of 0.8 L/min. The  $\text{N}_2$  flow rate was then changed to 0.2 L/min after heating began until the temperature in the furnace rose to 550 °C at the designated heating rate (15 K/min or 25 K/min). The furnace temperature was maintained for 40 min, and then heating ended; the same  $\text{N}_2$  flow rate was used to cool the furnace to the ambient temperature. During the pyrolysis process, the non-condensable gas was collected every 5 min for gas composition analysis using gas chromatograph (GC). At the end of the test, the condensed liquids and solid residues were collected directly and analyzed. The flow rate of the non-condensable gas was calculated by a mass balance.

Based on the results of different rubber-to-plastic blending ratios, the feedstock of the better rubber/plastics ratio was chosen as the optimized blending ratio to study the impact of the stalk additive on the co-pyrolysis process. The heating rate was another parameter studied, and the pyrolysis oils were analyzed by GC–MS. In most reference works, the heating rate of biomass pyrolysis for oil production is typically set at 15 K/min, 20 K/min, or 25 K/min [7–10].

The mass of the flask was measured before and after the test to calculate the mass of the pyrolysis oil. The ratio of the pyrolysis oil to the feedstock was then calculated to obtain the yield of extraction  $w_1$ . The oil heating value  $Q_1$  was measured using an oxygen bomb calorimeter. The yield of solid residues ( $w_s$ ) and the heating value of the residue ( $Q_s$ ) were obtained using the same method. The primary performance parameter discussed for the pyrolysis test is the pyrolysis oil yield. The other parameter of the feedstock-to-oil energy conversion efficiency (FOECE,  $\gamma_1$ ) is defined as the ratio of pyrolysis oil energy to the total energy of the feedstock, which is calculated using Eq. (1).

$$\gamma_1 = \frac{Q_1 \times w_1}{Q_R \times W_R + Q_P \times W_P + Q_S \times W_S} \times 100\% \quad (1)$$

where  $Q_R$ ,  $Q_P$ , and  $Q_S$  represent the heating values of rubber, plastics, and stalk;  $W_R$ ,  $W_P$ , and  $W_S$  are the mass fractions of the rubber, plastics, and stalk, respectively.

### 2.4. Analysis facilities

The collected liquid was centrifuged to separate the oil from the aqueous phase, and the residual moisture content in the oil was measured by a Trace Moisture Meter (SF101 type, China). The kinematic viscosity was measured under 40 °C using a kinematic viscosity meter (SYD-265C, China). The net heating values of the feedstocks, pyrolysis oils, and solid residues were measured using a fast calorimeter (WELL8000 Type). The element contents of the feedstocks and pyrolysis oil were measured using an elemental analyzer (Vario EL III).

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