



Comparative studies on catalytic and non-catalytic co-gasification of rubber seed shell and high density polyethylene mixtures



Bridgid Lai Fui Chin^a, Suzana Yusup^{a,*}, Ahmed Al Shoaibi^b, Pravin Kannan^b, Chandrasekar Srinivasakannan^b, Shaharin Anwar Sulaiman^c

^a Biomass Processing Lab, Centre for Biofuel and Biochemical Research, Green Technology MOR, Department of Chemical Engineering, Universiti Teknologi PETRONAS, Bandar Seri Iskandar, Tronoh 31750, Malaysia

^b Department of Chemical Engineering, The Petroleum Institute, P.O. Box 2533, Abu Dhabi, United Arab Emirates

^c Department of Mechanical Engineering, Universiti Teknologi PETRONAS, Bandar Seri Iskandar, Tronoh 31750, Malaysia

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ABSTRACT

The thermal degradation behavior of rubber seed shell, high density polyethylene waste, and the binary mixtures of high density polyethylene and rubber seed shell (0.2:0.8 weight ratio) are compared in pyrolysis, gasification, and catalytic gasification process at different heating rate of 10, 20, 30 and 50 K min⁻¹ in temperature range of 323–1173 K using thermogravimetric analysis equipment. It is observed that there are one, two, and three stages of decomposition occurring in high density polyethylene, rubber seed shell, and the binary mixtures respectively regardless of the process involved. The activation energies, E_A and pre-exponential factor, A are generated using one step integral method based on first order rate of reaction. Besides that, the synergistic effect on the binary mixtures is also investigated for the three different processes involved. It is found that the E_A and A values for the binary mixtures are lower than the pure high density polyethylene but comparable to pure rubber seed shell regardless of the processes involved. In addition, it is also observed that the E_A and A values are slightly lower when catalyst is added in the binary mixtures compared to the absence of catalyst in the gasification process.

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1. Introduction

Plastic waste is not only said to be one of the major constituents of municipal solid waste (MSW) but is also considered as one of the most harmful components in MSW due to its non-biodegradable properties which is inert to microbial degradation (Aboulkas et al., 2007; Kumar et al., 2011). From the statistics, polyethylene (PE) is reported to be the highest type of plastic consumption amongst the other polyolefins which takes about 33% of the world plastic consumption in year 2007 (CIPET, 2012). Among the polyethylene types, high density polyethylene (HDPE) is said to provide a major share of ethylene consumption structure recently with an evidence increased in world demand up to 4.4% in year 2009 (Kumar et al., 2011). HDPE possess linear structure with repeating unit of $-(CH_2-CH_2)-$, and contains the least degree of branching compared to other types of PE, which gives an advantage for the larger molecules in HDPE to be easily broken down into smaller

molecules with different chain length through the random-chain scission when heat is applied (Kumar et al., 2011). Finding an alternative method for effective plastic recycling continues to be a major barrier faced globally in replacement of conventional methods specifically incineration and land filling which is losing its significance due to its low operational cost effectiveness and descending credibility towards the environment (Aznar et al., 2006; Mumtaz et al., 2010). In solving the aforementioned problems, co-pyrolysis and co-gasification of plastic and biomass wastes is said to have gained considerable interest recently as it does not only subject to resolve the plastic waste disposal problems but could be an alternative source of renewable energy which could potentially convert these wastes to useful clean hydrocarbon fuel in the substitution of fossil fuels which is becoming increasingly scarce (Bosmans et al., 2013; Marin et al., 2002; Pinto et al., 2002).

Most publications report on the thermal degradation behavior and kinetic parameters for individual component of MSW and binary mixtures of plastic and biomass waste under co-pyrolysis and co-gasification condition. In contrast, no studies have been dedicated to catalytic co-gasification of binary mixtures of plastic and

* Corresponding author. Tel.: +60 5 368 7642; fax: +60 5 368 8205.

E-mail addresses: drsuzana_yusuf@petronas.com.my, drsuzana.yusuf.utp@gmail.com (S. Yusup).

biomass waste using thermogravimetric analysis (TGA). Saha and Ghoshal (2006) investigated on the non-isothermal and isothermal decompositions of polyethylene sample from polypack under nitrogen atmosphere of 40–50 mL min⁻¹ for temperature range of 303–873 K at different heating rates of 10, 15, 20 and 25 K min⁻¹. The activation energy, E_A dependency on conversion, x is compared with literature data and it is reported that the optimum activation energy, E_A dependencies of non-isothermal data at 15 K min⁻¹ heating rate are best predicted using the isothermal data. Aboulkas et al. (2008a) studied the thermal decomposition of co-pyrolysis olive residue and polypropylene mixtures in 1:1 ratio in a nitrogen atmosphere of 60 mL min⁻¹ for a range of temperature between 300 and 975 K at different heating rates of 2, 10, 20, and 50 K min⁻¹ and using Friedman isoconversional method to evaluate the decomposition kinetics. It is found that the maximum degradation temperature of the binary mixture is higher compared to the individual components alone. Thus, an increase in thermal stability is expected. It is also found that the presence of polypropylene does not significantly changed the apparent E_A of thermal decomposition of olive residue however the E_A value for polypropylene in the mixture is higher than that of the pure components. Chin et al. (2013a) investigated on the co-pyrolysis of rubber seed shell (RSS) with HDPE (0.8:0.2 weight ratio) in an argon atmosphere of 100 mL min⁻¹ for a temperature range of 323–1173 K at different heating rates of 10, 20, 30 and 50 K min⁻¹ and the kinetic parameters are determined using one step integral method. It is found that the E_A and pre-exponential factor (A) values for the binary mixture are lower than pure HDPE but slightly higher compared to pure RSS. The estimated E_A values for RSS, HDPE, and binary mixtures of RSS and HDPE are in the range of 46.94–63.21, 242.13–278.14, and 49.14–83.11 kJ mol⁻¹ respectively. Meanwhile, the estimated A for RSS, HDPE, and binary mixtures of RSS and HDPE are in the range of 3.20×10^6 – 1.31×10^8 , 8.30×10^{18} – 1.05×10^{22} , and 3.47×10^6 – 6.78×10^8 min⁻¹ respectively. It is also reported that the remaining residue increases with an increment of heating rate regardless the type of samples used. Chin et al. (2013b) studied the gasification kinetics of RSS, HDPE, and their mixtures (20/80 and 40/60 weight ratio of HDPE/RSS) at heating rate of 10 K min⁻¹ in the temperature range of 323–1173 K with argon gas supplied at flowrate of 100 mL min⁻¹ and steam injected at flowrate of 300 μ L h⁻¹ into TGA using Criado and Coats-Redfern methods. The predicted activation energy for RSS, HDPE, HDPE/RSS mixture (20/80 weight ratio), and HDPE/RSS mixture (40/60 weight ratio) are 385, 637, 177, and 175 kJ mol⁻¹ respectively. Chin et al. (2013c) examined the thermal decomposition behavior, degradation temperature, and maximum degradation rate of RSS, HDPE, and binary mixture of RSS and HDPE in the weight ratio of 20/80 (HDPE/RSS) at different heating rates of 10, 20, 30, and 50 K min⁻¹ within temperature range of 323–1173 K under non-isothermal gasification conditions with injected steam at flowrate of 300 μ L h⁻¹. The remaining solid residue after the gasification process for RSS, HDPE, and binary mixture of RSS and HDPE respectively are in the range of 11.9–15.3, 0.4–2.8, and 7.3–12.2 wt%. The maximum degradation rate achieved by the RSS, HDPE, and binary mixture of RSS and HDPE respectively are in the range of 7.9–43.7, 26.7–116.7, and 6.2–32.6 wt% min⁻¹.

In this present work, a comparison of the thermal degradation and kinetic parameters such as the activation energy (E_A) and pre-exponential factor (A) of RSS, HDPE waste, and the HDPE/RSS mixture (0.2:0.8 weight ratio) in pyrolysis, gasification, and catalytic gasification process using thermogravimetric analysis (TGA) approach at different heating rate of 10, 20, 30 and 50 K min⁻¹ in the temperature range of 323–1173 K using one step integral method are investigated. In addition, the synergistic effects on the binary mixture of RSS and HDPE are examined.

Table 1

Characteristics of the rubber seed shells (RSS) and high density polyethylene (HDPE).

Sample	Ultimate analysis (wt%, dry ash basis)					Proximate analysis (wt%, dry basis)			
	C	H	N	S	O ^a	MC	VM	FC ^a	Ash
RSS	44.31	4.38	0.51	0.13	50.67	8.59	80.98	6.62	3.81
HDPE	81.45	12.06	0.34	0.79	5.36	0.00	99.46	0.00	0.34

^a By difference.

2. Experimental

2.1. Materials and sample preparation

The raw materials used in this work are RSS from Vegpro Trading, Malaysia and HDPE plastic from Shen Foong Plastic Industries Sdn Bhd, Malaysia. These materials are ground and sieved to a particle size ranging from 0 to 710 μ m. Homogenized HDPE/RSS blends in a weight ratio of 0.2/0.8 are prepared. The characteristics of the materials used in this study are presented in Table 1. The ultimate analysis and proximate analysis of the HDPE and RSS are conducted in LECO CHNS-932 elemental analyzer and thermogravimetry analyzer EXSTAR TG/DTA 6300 (Seiko Instrument Inc.) respectively. A commercial Nickel powder (index number 028-002-01-4, Merck) is selected as a catalyst to be investigated in the catalytic gasification process for comparison of pyrolysis and gasification behavior of HDPE/RSS binary mixture. The amount of Nickel powder used in the present study is 10% of the total weight of the HDPE/RSS binary mixture. The particle size of the Nickel powder is in the range of \sim 10 μ m and the properties of the catalyst is presented in Table 2. The field emission scanning electron microscopic (FESEM) images of the Nickel catalyst in 7000 and 10,000 \times magnification determined using Zeiss Supra 55 VP are displayed in Fig. 1 to depict the surface physical morphology of Nickel powder. It is observed that the surface morphology of Nickel powder is a flower-like shape with nanotips outspreading from the cores which are similar to the work reported by Bai et al. (2008) and Kim et al. (2004).

2.2. Experimental apparatus and procedure

The experiments are performed using thermogravimetry analyzer EXSTAR TG/DTA 6300 (Seiko Instrument Inc.) as illustrated in Fig. 2. Approximately 5 mg of sample is placed on a ceramic crucible in TGA under an inert atmosphere of argon. A flow rate of 100 mL min⁻¹ of argon gas is fed into the system for 20 min at a temperature of 323 K. Subsequently, all samples are heated from 323 K to 1173 K at respective heating rates and temperatures are kept constant for 10 min. During heating, the TGA is used to measure mass variation of the materials and furnace temperature. Thermogravimetric curves are obtained at four heating rates (10, 20, 30, and 50 K min⁻¹) within the temperature range of 323–1173 K. Steam is generated by a superheater at 383 K and is injected into the TGA at a flowrate of 300 μ L h⁻¹ when the temperature inside the TGA equipment reached to 383 K to avoid any occurrence

Table 2

Properties of Nickel powder.

<i>XRF analysis</i>	
Ni (wt%)	96.79
Impurities (wt%)	3.21
<i>Surface properties</i>	
Mean pore size (nm)	6.2
Pore volume (BJH) (cm ³ g ⁻¹)	0.0016
BET surface area (m ² g ⁻¹)	0.78

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