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Catalytic co-pyrolysis of waste vegetable oil and high density polyethylene for hydrocarbon fuel production

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ABSTRACT

In this study, a ZrO₂-based polycrystalline ceramic foam catalyst was prepared and used in catalytic co-pyrolysis of waste vegetable oil and high density polyethylene (HDPE) for hydrocarbon fuel production. The effects of pyrolysis temperature, catalyst dosage, and HDPE to waste vegetable oil ratio on the product distribution and hydrocarbon fuel composition were examined. Experimental results indicate that the maximum hydrocarbon fuel yield of 63.1 wt. % was obtained at 430 °C, and the oxygenates were rarely detected in the hydrocarbon fuel. The hydrocarbon fuel yield increased when the catalyst was used. At the catalyst dosage of 15 wt.%, the proportion of alkanes in the hydrocarbon fuel reached 97.85 wt.%, which greatly simplified the fuel composition and improved the fuel quality. With the augment of HDPE to waste vegetable oil ratio, the hydrocarbon fuel yield monotonously increased. At the HDPE to waste vegetable oil ratio of 1:1, the maximum proportion (97.85 wt.%) of alkanes was obtained. Moreover, the properties of hydrocarbon fuel were superior to biodiesel and 0# diesel due to higher calorific value, better low-temperature low fluidity, and lower density and viscosity.

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1. Introduction

Since the last century, the increasing energy crisis has initiated many studies on renewable energy as an alternative to traditional fossil fuels. CO₂ from biomass combustion was released into the atmosphere and then returned to the biomass under the action of photosynthesis. The circulation pathway does not cause extra burden to environment (McKendry, 2002). Therefore, biomass utilization technologies including fast pyrolysis (Bridgwater, 2012) and hydrothermal processing (Ross et al., 2010) are considered very promising in the future biorefinery. In China, there is much waste vegetable oil produced from catering industry and food factory every year, which causes increasing disposal problems and environmental issues. Since waste vegetable oil is mainly composed of triglyceride, it is widely used to produce biodiesel via transesterification (Lopresto et al., 2015; Ghoreishi and

Moein, 2013). However, the application of biodiesel is limited by its low cetane number and calorific value. An efficient and low-cost pathway for conversion of waste vegetable oil to high-quality fuels is still an important research topic.

Fast pyrolysis is regarded as an alternative way to produce fuels from triglyceride, which is a relatively technically mature process. As a thermochemical conversion technology, biomass fast pyrolysis at high heating rate in inert atmospheres can yield liquid products, bio-char and gaseous products (Bridgwater, 2012; Butler et al., 2011). Recently, several studies demonstrated that pyrolyzing triglyceride can obtain superior quality bio-oil with the advantages of low viscosity, low freezing point and cold filter plugging point, and high cetane number (Wang et al., 2012; Abnisa et al., 2013). Wiggers et al. (2009) investigated the continuous pyrolysis of soybean oil at temperatures ranging from 450 °C to 600 °C in a pilot plant and obtained biofuels similar to fossil fuels. Results of thermal analysis indicate that it is possible to use the liquid products as an energy source for the process. Ben Hassen-Trabelsi et al. (2014) pyrolyzed several animal fatty wastes under nitrogen in a laboratory scale fixed-bed reactor. When the pyrolysis temperature and heating rate were set at 500 °C and 5 °C/min, respectively, the maximum yield of bio-oil was achieved. The bio-oil consisted

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of hydrocarbons, carboxylic acids, aldehydes, ketones, esters, etc., and was suitable to be used as an engine fuel. Thus, fast pyrolysis of waste vegetable oil offers an alternative pathway to produce renewable liquid products used for fuels or chemicals. It is worth noting that the bio-oil obtained from fast pyrolysis cannot be directly applied to machines unless it is upgraded.

Abundant waste plastics from municipal solid waste and agricultural industry cannot be degraded, making their post-treatment and recycling difficult. Therefore, finding an appropriate disposal route to take advantage of these abundant waste plastics as a source of energy or chemicals becomes a great challenge in the world. In recent years, the co-pyrolysis technique has attracted much attention because the process produces high-quality pyrolysis oil. In addition, many studies selected plastics as one of the co-feeds to improve the quality and yield of bio-oil. High density polyethylene can act as a hydrogen donor in the co-pyrolysis with biomass which usually has low hydrogen content (Aboulikas et al., 2012). The synergetic effect between the oil shale and high density polyethylene leads to an increase in oil yield and a decrease in the oxygen content of shale oil. Önal et al. (2012) conducted co-pyrolysis of potato skin and high density polyethylene, and found that the addition of high density polyethylene improved the liquid products in terms of both quality and quantity.

Proper catalyst plays an important role in conducting thermochemical reactions selectively. Along with the development of theories, catalyst preparation techniques, and study on traditional homogeneous catalytic reactions, ZrO₂-based metal oxide catalyst has become a research focus due to its excellent catalytic performance for biomass catalytic cracking and polymerization (Lan et al., 2015; Takase et al., 2014; Gürbüz et al., 2010; Kunkes et al., 2009). The CeO₂-ZrO₂-Al₂O₃ catalyst prepared by coprecipitation method presented superior textural, structural, morphological and reduction properties along with enhanced three-way catalytic performance (Lan et al., 2015). Duan et al. (2009) pointed out that the binary oxides of SiO₂, TiO₂, ZrO₂ modified γ -Al₂O₃ showed promising properties in catalysis processes. The Pt/ZrO₂-TiO₂-Al₂O₃ catalyst had good catalytic cracking activity due to its large amounts of strong acid sites, large surface area and pore volume, which evidently improved the gas yield and heat sink of catalytic cracking of RP-3 (Jiao et al., 2015). Dumesic and his co-workers reported that ketonization and aldol condensation/hydrogenation were two important C–C coupling reactions for upgrading mono-functional oxygenated compounds for transportation fuel applications (Gürbüz et al., 2010; Kunkes et al., 2009). Carboxylic acids in the reactions can be ketonized with nearly 100% conversion over ceria-zirconia mixed oxide catalyst (Kunkes et al., 2008). The C–C coupling reactions of ketones via aldol condensation/hydrogenation over Pd/CeZrO_x and CeZrO_x catalysts were studied, finding that ceria-zirconia mixed oxide catalyst contributed to the conversion of ketones into corresponding fuel-grade alkanes via aldol condensation/hydrogenation (Kunkes et al., 2009). In this regard, ZrO₂-based catalysts have high selectivity of fuel-grade hydrocarbons in comparison to other catalysts. Thus, based on the potential catalytic performance of zirconia oxide as catalyst according to literatures, a ZrO₂-based polycrystalline ceramic foam catalyst was synthesized in this study using ceramic foam research methods and preparation techniques to improve hydrocarbon fuel properties (Chen et al., 2013; Thompson et al., 2013).

In this study, catalytic co-pyrolysis of waste vegetable oil and HDPE was carried out with a ZrO₂-based polycrystalline ceramic foam catalyst for hydrocarbon fuel production under different conditions. This process can convert wastes into high value-added products without pollution. Moreover, the obtained liquid products were prospective to be substitute for petrochemical resources. The ZrO₂-based polycrystalline ceramic foam catalyst

was characterized using temperature-programmed desorption (TPD) of CO₂ and NH₃, X-ray diffraction (XRD), and BET surface area measurements. The effects of co-pyrolysis temperature, catalyst dosage, and HDPE to waste vegetable oil ratio on the product distribution and hydrocarbon fuel composition were investigated.

2. Materials and methods

2.1. Materials

The pyrolysis feedstock was waste vegetable oil and HDPE. The waste vegetable oil, non-edible sunflower oil undergoing spoilage, was provided by student canteen of Nanchang University in Nanchang, Jiangxi Province, China. High density polyethylene plastics were obtained from solid waste disposal plant in Nanchang. Prior to use, HDPE was mechanically pulverized and sifted through a 40-mesh sieve. ZrO₂, Al₂O₃, TiO₂, NaCl, NaHCO₃ (AR) were purchased from Aladdin (China) and not treated prior to use. The acidic silica sol was from Qingdao Jiyida silica reagent Corporation (China). The basic physico-chemical characteristics of waste vegetable oil and HDPE are shown in Table 1. Waste vegetable oil was dried in a vacuum freeze drier at –80 °C for 48 h to measure the moisture content. The CHONS contents of waste vegetable oil and HDPE were determined via ECS 4010 CHONS elemental analyzer. The fatty acids composition of waste vegetable oil was measured using GC-MS. Before GC-MS analysis, fatty acids were turned to fatty acids methyl esters by means of methylation. The GC-MS analyses and conditions were presented in Section 2.4.

2.2. Catalyst preparation and characterization

2.2.1. Catalyst preparation

A mixture of ZrO₂ (75.8%), Al₂O₃ (8.4%), and TiO₂ (4.2%) was ball-milled for 24 h in the globe mill. NaCl and NaHCO₃, used as pore-forming agent and acidic silica sol serving as the binder, were added to the mixture. The mixture was uniformly mixed by a comminutor and subsequently granulated with the diameter of 4 mm. The catalyst was dried at 120 °C for 12 h in the oven. After being cooled down to the room temperature, the catalyst was calcined at 500 °C in a muffle furnace for 1 h to remove the low temperature pore-forming agents and then heated to 1550 °C rapidly to calcine continuously for 2 h. In addition, the high temperature pore-forming agents were completely evaporated after being heated to a specified temperature rapidly. The mixture was finally cooled down to obtain the ZrO₂-based polycrystalline ceramic foam catalyst.

2.2.2. Catalyst characterization

The BET surface area was measured by nitrogen absorption using a Kubo-X1000 multi-functional micropore analyzer. Pore

Table 1
Characteristics of waste vegetable oil and HDPE.

	Waste vegetable oil	HDPE
Moisture content (wt.%)	0.16	Not detected
Elemental composition (wt.%)		
C	78.14	84.5
H	9.27	14.7
N	0.10	0
O	12.36	0.8
S	0.13	0
Fatty acid composition		
Palmitic acid (C16:0)	7.9	
Stearic acid (C18:0)	4.2	
Oleic acid (C18:1)	36.6	
Linoleic acid (C18:2)	51.3	

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