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Hydrocracking of palm oil to jet biofuel over different zeolites

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ABSTRACT

The aim of this study is to develop zeolite catalysts to produce jet biofuel from palm oil. Different Ni-loaded zeolites were tested as catalysts in the conversion of palm oil into jet biofuels with high alkane and low arene content. Five zeolite catalyst, were tested: Ni/SAPO-34, Ni/MCM-41, Ni/HY, Ni/SAPO-11 and Ni/Hbeta. Characterization was performed by X-ray diffraction, NH₃-temperature-programmed desorption. The Ni/SAPO-34 catalyst exhibited the highest alkane selectivity (65%) and lowest arene selectivity (11%). The jet biofuel yield under catalysis increased from 21.1 to 42.0% when the reaction temperature was increased from 370 to 390 °C. The alkane content of the jet biofuel increased from 71 to 80% and the arene content decreased from 29 to 20%, when the weight ratio of Si/Al in the Ni/HY catalyst was reduced from 11 to 5, but the levels were almost unchanged as the nickel content was increased from 5 to 20%.

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Introduction

The aviation industry is in a phase of development triggered by declining petroleum resources, increased crude oil prices and stringent environment policies. Jet fuel consumption increased from 189.1 million gallons per day in 2001 to 198.3 million gallons per day in 2008 [9,10], and the use of bio-based jet fuel could reduce greenhouse gas emissions by up to 89% [8]. Petroleum-based jet fuel contains C₈–C₁₆ alkanes, cycloalkanes, olefins, and arenes. The contents of mono-arene and polycyclic arene in jet fuel are limited because of the high heating values [3]. However, jet fuel with insufficient arene content causes poor lubrication. Bio-based jet fuel can be produced through the hydrotreatment of plant oil, Fischer–Tropsch synthesis of biomass, and pyrolysis. However, both Fischer–Tropsch synthesis and biomass pyrolysis are

complex processes. Plant oil hydrotreatment offers a simpler approach to the production of jet fuel.

Wu et al. [16] reported diesel range alkane production through rotating gliding arc discharge. Many researchers have investigated plant oil hydrotreatment for diesel fraction (C₁₆–C₂₃) production [9–11,13,14], but there have been few studies of jet fuel production from plant oil. Jet fuel can be produced from plant-derived fatty acids through deoxygenation and carbon chain cracking. Verma et al. [15] demonstrated jet fuel production from jatropha oil and algae lipids via a one-step process. However, the jet fuel produced contained only small amounts (1%) of the aromatics that make up 25% by volume of petroleum jet fuel and that are necessary for proper lubrication and sealing [1]. Cheng et al. [4] demonstrated the production of jet fuel from soybean oil, but soybean oil is edible and is therefore unsuitable for use as an

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industrial product. Moreover, jet fuel converted from soybean oil via zeolite HY catalysis contains a very high proportion of arene [4]. The optimal proportion of arene in jet biofuel should be in the range of 10–15%. Robota et al. [12] demonstrated jet biofuel production from algal lipids. However, the yield of jet biofuel is very low.

Palm oil is an economically important plant that is traded worldwide. In 2005, the total world supply of palm oil was 32.65 million tons. In this study, palm oil was used as a feedstock for jet fuel production, with Ni supported on zeolites with different pore structures used as the catalyst. Zeolite SAPO-34 exhibited the lowest arene selectivity and the highest alkane selectivity. Optimization of the reaction temperature was also investigated.

Materials and methods

Preparation of catalysts

The analytical standard $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ($\geq 98.0\%$ purity) used in the experiments was purchased from Sinopharm Chemical Reagent Co. Ltd, Shanghai, China. Zeolites HY, Hbeta, MCM-41, SAPO-34 and SAPO-11 were purchased from the Catalyst Plant of Nankai University. Ni clusters supported on zeolite catalysts were prepared using the wetness impregnation method, and a similar procedure was used to synthesize the Ni (10 wt%)/zeolite catalysts. For example, Ni (10 wt%)/zeolite SAPO-34 catalyst was synthesized as follows: 2.97 g $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ was dissolved in 10 ml of deionized water. Then, 5.4 g of zeolite SAPO-34 was added to the solution. The mixture was stirred for 4 h at ambient temperature, and then dried in an oven at 70 °C for 8 h. The catalyst was calcined in air at 550 °C (heating rate = 5 °C/min) for 4 h and reduced in hydrogen (flow rate = 350 ml/min) at 500 °C (heating rate = 4 °C/min) for a total of 4 h.

Characterization of catalysts

X-ray diffraction (XRD) patterns of the catalysts were obtained using an X'Pert PRO MPD diffractometer (PANalytical) operated at 40 kV and 40 mA with $\text{Cu K}\alpha$ radiation. NH_3 -temperature-programmed desorption (NH_3 -TPD) experiments were performed using a Micrometrics AutoChem II 2920 system.

Preparation of jet biofuel

The palm oil used in the experiments was purchased from HuanShuYuan Company, Shanghai, China. The jet biofuel was prepared in a 500 ml batch reactor (Parr Instrument Company 4500) equipped with a mechanical stirrer. In a typical run, 100 ml of palm oil and catalyst at a 20:1 mass ratio were loaded in the reactor. The reactor was then sealed and filled with hydrogen to control the pressure and temperature. The reaction was performed with a stirring speed of 500 rpm at 370–410 °C for 8 h. The liquid and solid products were separated via centrifugation after the reaction. The weight of the liquid products was measured on a balance, and the liquid composition was analyzed using Gas Chromatography Mass Spectrometry (GC–MS).

Analysis of liquid products

The liquid product samples were diluted to a ratio of 1:10 in chloroform and were analyzed using an Agilent 6890N GC/5975B MSD equipped with an HP-5 capillary column. The injection temperature was set to 320 °C. A high injection port temperature was used to ensure reliable and direct quantification of the fatty acids and triglycerides without chemical derivatization [7,2]. The column temperature was initially increased from 30 to 80 °C at a rate of 2 °C/min, then increased to 300 °C at a rate of 10 °C/min, and finally maintained at 300 °C for 20 min. The GC–MS results were quantified using the peak area normalization method based on the peak area percentages of the identified components. All measurements were conducted in triplicate. Mean values and standard deviation were recorded. Yield, selectivity, and conversion were defined as follows:

$$\text{Yield} = (\text{mass of the product}/\text{mass of soybean oil}) \times 100\%.$$

$$\text{Selectivity} = (\text{mass of the product}/\text{mass of total products}) \times 100\%.$$

$$\text{Conversion} = (\text{mass of converted palm oil}/\text{mass of palm oil}) \times 100\%$$

Results and discussion

Catalyst characterization

The XRD patterns of the Ni/SAPO-34, Ni/SAPO-11, Ni/Hbeta, Ni/HY, and Ni/MCM-41 catalysts are shown in Fig. 1(a). The XRD patterns of the SAPO-34, SAPO-11, Hbeta, and HY were consistent with the LTA, AEL, BEA, and FAU structures of these zeolites. Because the Ni was supported on five different zeolites, the diffraction intensities responsible for the Ni crystallites were clear. The diffraction intensity of the Ni crystallites supported on zeolite Hbeta was weaker than that of the other samples, suggesting that the Ni crystallites were smaller than those of the other samples.

Fig. 1(b) shows the NH_3 -TPD profiles of the Ni/SAPO-34, Ni/SAPO-11, Ni/Hbeta, Ni/HY, and Ni/MCM-41 catalysts. For the Ni/SAPO-11 catalyst, NH_3 desorption peaks at 177 and 342 °C were observed, suggesting the presence of weak and medium acidic sites on the catalyst surface. In the case the Ni/MCM-41 catalyst, an NH_3 desorption peak at 345 °C was observed, indicating the presence of a medium acidic site. For the Ni/HY catalyst, an NH_3 desorption peak at 180 °C was observed, indicating the presence of a weak acidic site on the catalyst surface. For the Ni/SAPO-34 catalyst, NH_3 desorption peaks at 170 and 320 °C were observed, indicating the presence of weak and medium acidic sites. For the Ni/Hbeta catalyst, an NH_3 desorption peak at 320 °C was observed, indicating the presence of a medium acidic site on the catalyst surface.

Conversion of palm oil into jet biofuel using Ni/HY catalysts of different Si/Al ratios and Ni contents

In our previous research, zeolite HY was used as a catalyst when converting soybean oil into jet biofuel. In this study, zeolite HY with a range of Ni contents and Si/Al ratios was

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