Appl Biochem Biotechnol (2010) 162:1725–1736
DOI 10.1007/s12010-010-8953-9

Biodiesel Preparation from *Jatropha curcas* Oil Catalyzed by Hydrotalcite Loaded With K_2CO_3

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Received: 27 December 2009 / Accepted: 15 March 2010 /

Abstract This paper discusses the synthesis of biodiesel catalyzed by solid base of K_2CO_3/HT using *Jatropha curcas* oil as feedstock. Mg–Al hydrotalcite was prepared using co-precipitation methods, in which the molar ratio of Mg to Al was 3:1. After calcined at 600 °C for 3 h, the Mg–Al hydrotalcite and K_2CO_3 were grinded and mixed according to certain mass ratios, in which some water was added. The mixture was dried at 65 °C, and after that it was calcined at 600 °C for 3 h. Then, this Mg–Al hydrotalcite loaded with potassium carbonate was obtained and used as catalyst in the experiments. Analyses of XRD and SEM characterizations for catalyst showed the metal oxides formed in the process of calcination brought about excellent catalysis effect. In order to achieve the optimal technical reaction condition, five impact factors were also investigated in the experiments, which were mass ratio, molar ratio, reaction temperature, catalyst amount and reaction time. Under the best condition, the biodiesel yield could reach up to 96%.

Keywords Biodiesel · Transesterification · Hydrotalcite · Solid base · *Jatropha curcas* oil

Introduction

With the rapid development of human society and economy, demand of energy is becoming more and more tremendous. On the contrary, the petroleum, one of the most important energy resources, becomes less and less and its reserve is not unlimited. Thus, the shortage of fossil diesel will become a great challenge for human beings. Owning to the importance of diesel, how to find an ideal and practical substitute of diesel is drawing much attention from a lot of researchers and scientists all over the world. [1]. Biodiesel, which is called fatty acid methyl esters (FAMEs) chemically, is a promising alternative of fossil diesel [2–4]. It is usually

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obtained by transesterification reaction between alcohols and vegetable oils or animal fats. Compared with fossil diesel, biodiesel has many advantages, such as lower emission of SO*x*, more sufficient combustion, benign biodegeneration, and extensive source of raw materials [5]. What is more, biodiesel's combustion performance is as good as common diesel's or even better in some aspects. Therefore, research on biodiesel is very helpful to solve the energy crisis problem and to promote the sustainable development of mankind.

Nearly all the common oils and fats have been studied to prepare biodiesel by researchers [6–9]. Nevertheless, most of those materials are edible or/and expensive and the over use of them would cause or aggravate the crisis of cereals and edible oils. On the other hand, it is unpractical to use the over-expensive materials in actual production of biodiesel. *Jatropha curcas* oil, a promising and inedible source of biodiesel, has drawn the attention of many researchers who they had made some progress [10, 11]. Because of these advantages, curcas oil was selected as material oil in these experiments. Catalyst is another crucial influence factor in biodiesel preparation. Although acidic catalysts were used by many researchers, the serious erosion of manufacture equipment and troublesome post-treatment problems during the experiments proved that acidic catalysts are not workable for mass production [12–14]. Highly efficient lipase and enzymatic catalysts were also used by some investigators [15–20]. However, the high price and trenchant reaction conditions restrict the application of lipase catalyst in large-scale production. Oil reacted with supercritical methanol without any catalyst could be seen in some literatures, in which reaction conditions limited the production as well [21–29]. Biodiesel preparation under microwave condition has the dilemma of high energy consumption [30]. Lots of researchers chose solid bases as catalysts because of their particular advantages, such as low price, easy regeneration, high efficiency, effortless separation from crude product, and etc. [31–33].

Hydrotalcite (HT), showed as $Mg_{1-X}Al_X(OH)_2(CO_3)_{X/n}\cdot nH_2O$, was used as precursors and supporters, because of its strong alkalescence and double-layered structure [34–37]. Therefore, as a carrier, it was used in the preparation of biodiesel [38]. In this work, HT was used to prepare a strong basic catalyst which then was used to catalyze the trans-esterification reaction. HT was prepared with magnesium and aluminum salts, in which the molar ratio of Mg^{2+} to Al^{3+} was 3:1. Then, the HT was calcined at 600 °C. The calcined HT and K_2CO_3 were grinded adequately according to certain mass ratios with some water. After the mixture was kept at 65 °C to dry (about 10 h) and was calcined again, the catalyst was obtained. Technically, all the impact factors were investigated and the optimal condition was recorded. Under the optimal condition, yield of biodiesel could reach 96.69%. XRD patterns showed the excellent catalysis effect was brought by new metal oxides. SEM images showed the addition of potassium improved the performance of HT and intensified catalysis activity.

Experimental Section

Materials

Jatropha curcas oil was bought from Shuyang County, Jiangsu Province, China. Fatty acid composition of *Jatropha curcas* oil was showed in Table 1. Methanol, NaOH, Na_2CO_3, and K_2CO_3 were the products of Nanjing Ningshi Chemical Reagent Co., Ltd. Shanghai Zhenxin Reagent Plant supplied $Al(NO_3)_3\cdot 9H_2O$ and $Mg(NO_3)_2\cdot 6H_2O$.

All the reagents used in the experiments were analytical grade.

Table 1 Fatty acid composition of *Jatropha curcas* oil.

Components	Formula	Content (%)
Myristic acid	$C_{14}H_{28}O_2$	0–0.1
Palmitic acid	$C_{16}H_{32}O_2$	14.1–15.3
Stearic acid	$C_{18}H_{36}O_2$	3.7–9.8
Arachidic acid	$C_{20}H_{40}O_2$	0–0.3
Behenic acid	$C_{22}H_{44}O_2$	0–0.2
Oleic acid	$C_{18}H_{34}O_2$	34.3–45.8
Linoleic acid	$C_{18}H_{32}O_2$	29.0–44.2
Linolenic acid	$C_{18}H_{30}O_2$	0–0.3
Palmitoleic acid	$C_{16}H_{30}O_2$	0–1.3

Data from Gubitz et al. [39]

Catalyst Preparation

Co-precipitation method was used to prepare Mg–Al hydrotalcite (HT) in which the element ratio of Mg to Al was 3:1. Three solutions were prepared with deionized water first, which were marked solution A, B and C. Solution A was 400 mL, which was prepared with 0.36 mol magnesium nitrate and 0.12 mol aluminum nitrate. Solution B (200 mL) contained 0.6 mole NaOH, and solution C (200 mL) included 0.3 mole Na_2CO_3.

Solution A was added into a 1,000-mL four-neck-flask with thermostatic oil bath and vigorous stirring, keeping the oil temperature at 65 °C. Then, NaOH solution and Na_2CO_3 solution were added into the flask dropwise and respectively. After solutions B and C were dripped off, the precipitate was kept at 65 °C with vigorous mechanical stirring. After 24 h, the mixture was filtered and washed until the pH value was kept at 8–9.

After that, the mixture was kept at 65 °C to dry (about 24 h), then calcined at 600 °C for 3 h. K_2CO_3 and the calcined HT were blended and grinded adequately according to certain mass ratio with some deionized water, 1:10, 3:10, 5:10, 7:10, and 9:10. This mixture was kept at 65 °C for 24 h then calcined at 600 °C for 3 h.

The finished catalysts were labeled as 10%, 30%, 50%, 70% and 90% K_2CO_3/HT, respectively, according to the mass ratios.

Transesterification Reaction

Experiments of transesterification reaction were finished in a 250-mL four-neck flask reactor with rigorous electric stirring. Methanol was added into the flask and heated to setting temperature first. Then, *Jatropha curcas* oil and certain mass catalyst were also heated to the setting temperature and were added into flask. Once the reaction reached certain time, the temperature of reactor with mixture inside was decreased to normal temperature as soon as possible once the reaction was finished.

The mixture was filtered and collected into a flask. After some time, the filtered mixture was layered. The upper layer was mainly the mixture of fatty acid methyl esters, methanol and unreacted oil, and the lower layer was mainly glycerol and methanol because the glycerin was weightier. Therefore, the upper layer mixture/sample was sent to test and analyze the yield.

First, the impact of mass ratio was investigated while other influence factors were set invariable. Similarly, the next four factors to yield of biodiesel: molar ratio of methanol to

Jatropha curcas oil, reaction temperature, catalyst amount (mass percentage of *Jatropha curcas* oil), and reaction time were investigated one by one.

Analysis and Characterization

The yield of methyl ester of product was tested by gas chromatography (GC) provided with a flame ionization detector, employing a silica capillary column of 15 m long and diameter of 0.32 mm inside. The gas chromatography was supplied by Shanghai Ouhua Analysis Apparatus Factory, which number was GC-9160.

In GC patterns, because the same detector engendered different response values for different substances, two mass-equal substances brought about different peak areas. Therefore, the peak area could not be used as a factor to calculate the mass of a substance directly; some process must be needed first. Thus, to compute the mass of substance exactly, a factor was drawn into calculations, which was called correction factor.

In the GC patterns, peak area of all the fatty acid methyl esters peaks was set, a. Similarly, peak area of monoglyceride, peak area of diglyceride and peak area of triglyceride (oil) were set as b, c, and d. In addition, correction factors were set as f_a, f_b, f_c, f_d, respectively. Then the calculating equation was gotten as below,

$$\text{yield}\ (\%) = \frac{a \times f_a}{a \times f_a + b \times f_b + c \times f_c + d \times f_d} \tag{1}$$

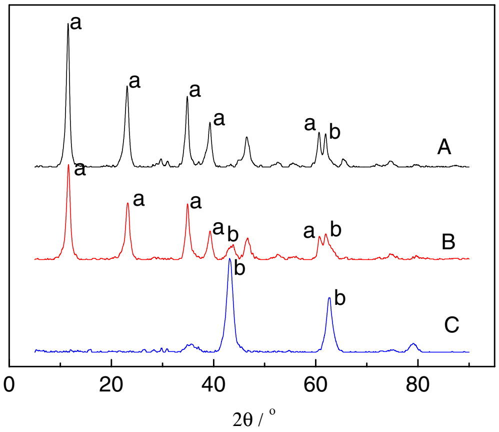
Catalyst was characterized with X-ray diffractions (XRD) and scanning electron microscope (SEM).

Results and Discussion

Catalyst Characterizations

X-ray diffraction patterns of calcined HT and catalysts with different load ratios are showed in Fig. 1, in which A, B and C are the patterns of pure HT, 10% K_2CO_3/HT, 50% K_2CO_3/HT, respectively. It is easy to see there are strong peaks existing at 11.8° 23.3°, 35.1°, 39.5°, 47.5°, 53.4°, and 61.1° in pattern A (peaks *a*), which are typical HT peaks and show the

Fig. 1 X-ray diffraction patterns of different HT and different catalysts. *a* calcined HT, *b* catalyst with 10% load ratio, *c* catalyst with 50% load ratio



double-layered structure of HT [34, 40]. Compared with pattern A, in the pattern B, the strengths of peaks *a* at the same positions are smaller. And there is a new peak showing at 43.2°, although its strength is not huge. Compared with patterns A, in pattern C, nearly all the peaks *a* disappear. However, there are two very strong peaks showing at 43.2° and 62.3° (peaks *b*). In terms of catalysis effect, 50% K_2CO_3/HT is more efficient than 10% K_2CO_3/HT. According to these three XRD patterns, it is thought that a new crystal is produced. The new crystal might be multiple metal oxides. Because 10% K_2CO_3/HT has less new crystal, the catalysis effect is not as splendid as the effect of 50% K_2CO_3/HT. Therefore, it is a good reason to explain why 50% K_2CO_3/HT has excellent catalysis effect to transesterification reaction of *Jatropha curcas* oil.

In Fig. 2, XRD patterns of used and unused 50% K_2CO_3/HT are showed. Both patterns C and D have similar peaks at 43.2° and 62.3° (peaks *b*). However, the strengths of the two peaks of used catalyst are weaker than the unused catalysts. Yet, main peaks sharps of used catalyst still exist. Thus, this shows that catalysis effect of used catalyst decreases a little, but it is still very strong in transesterification reaction. In this way, it is concluded that catalysis kept steady for a long time.

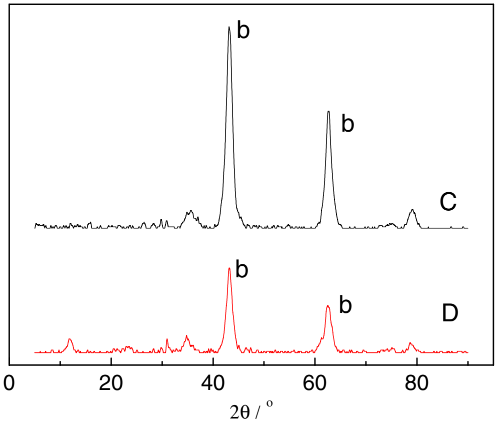
SEM images of Mg–Al HT, 50% K_2CO_3/HT and the used 50% K_2CO_3/HT were observed to determine the morphology and particle size distribution. As clearly shown in Fig. 3, the Mg–Al HT particle size distribution is not symmetric in the range of 2 to 60 μm (Fig. 3a). Layered structure of Mg–Al HT is also indicated by the thin flat crystals and the edges (Fig. 3a). However, owing to the calcination at 600°C, the layered structure of HT is not very excellent. It is clearly showed that the morphology of Fig. 3b's catalyst is different from the morphology of Fig. 3a. Generally, the particle size of 50% K_2CO_3/HT became larger than the Mg–Al HT. However, it could be seen the layered structure was optimized after potassium being loaded. Because of the benign layered structure, specific surface area increases accordingly.

Compared with Fig. 3b, no obvious difference is observed in Fig. 3c. The morphology and particle size distribution of catalyst keeps no changes. Therefore, it is proved again that the catalysis effect could keep steady for a long time.

Transesterification Reaction

Effect of reaction temperature on the yield of biodiesel is showed in Fig. 4. From 35 to 65 °C, biodiesel yield increased from 12.7% to 96.69% which was the highest value of all. However, there was a slight decrease from 65 to 75 °C where yield of biodiesel was

Fig. 2 X-ray diffraction patterns of used and unused catalysts. Both catalyst load ratios were 50%, *c* unused catalyst, *d* used five times catalyst



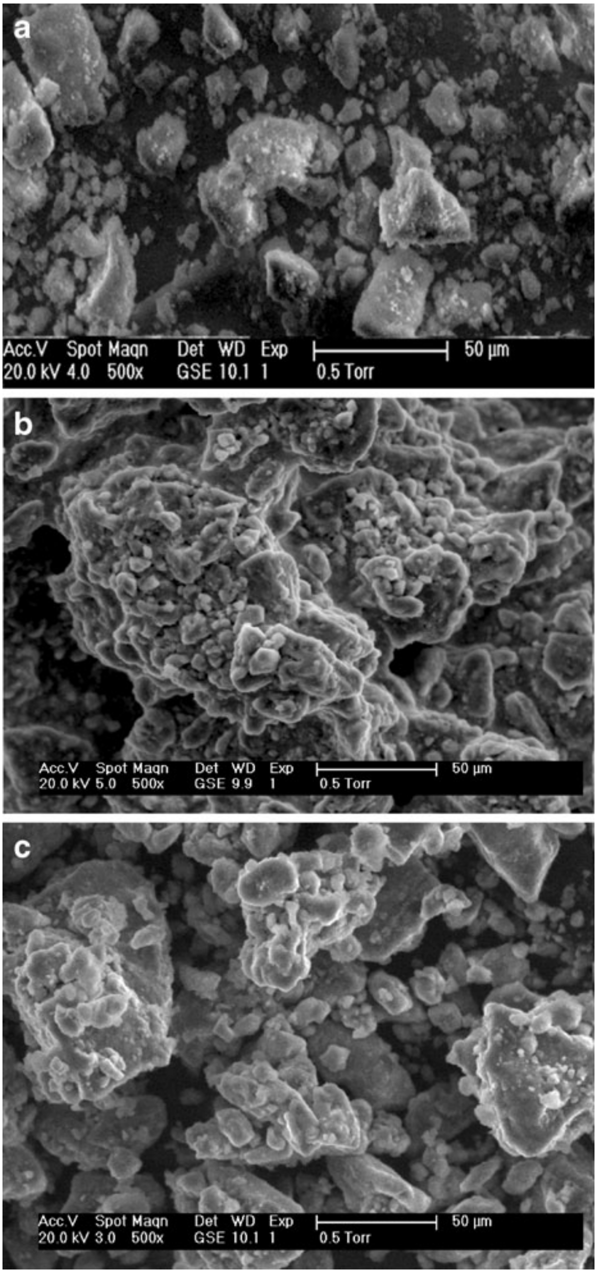
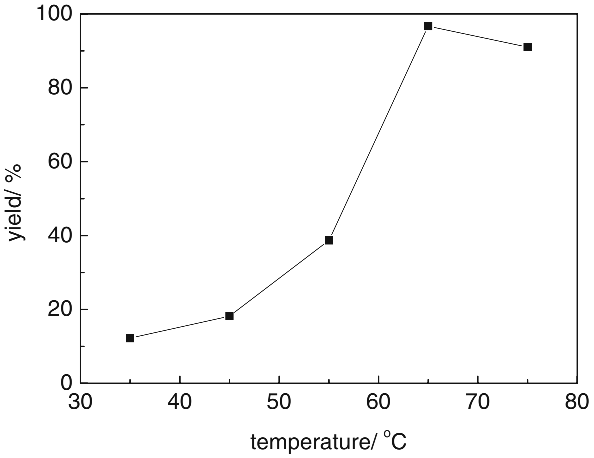


Fig. 3 SEM images of Mg–Al HT, 50% K_2CO_3/HT and used 50% K_2CO_3/HT. **a** Mg–Al HT, **b** 50% K_2CO_3/HT; **c** used five times 50% K_2CO_3/HT

90.01%. It is likely that higher temperature drove molecules of methanol and triglyceride more active, in which their collision probability and chance increased. Higher crash opportunities drove the transesterification reaction easier. Therefore, with the temperature increasing from 35 to 65 °C, the yield of biodiesel increased drastically.

Nonetheless, there is a little reduction when the reaction temperature was at 75 °C. It was thought which was caused by serious volatilization of methanol. Boiling point of

Fig. 4 Effect of reaction temperature on yield of FAMEs. Reaction condition: molar ratio was 12:1, catalyst amount was 3%, reaction time was 3 h, and mass ratio was 50%

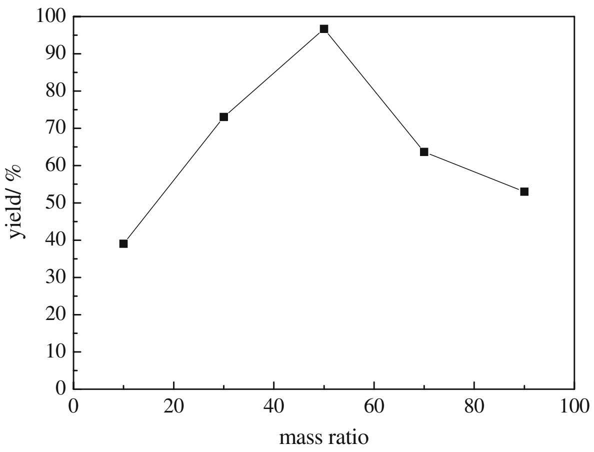


methanol was about 65 °C, over-high temperature caused methanol run out of liquid which lowered the reciprocal collision opportunities of oil and methanol molecules. Therefore, yield was lower than the value when the reaction temperature was 65 °C. Therefore, 65 °C was selected as the best reaction temperature.

In Fig. 5, effect of mass ratio of K_2CO_3 to HT on FAMEs yield is shown. Obviously, yield of FAMEs increases when mass ratio increased from 10% to 50% and decreases when mass ratio is from 50% to 90%. It could be the reason that when mass ratio is 10%, the active component is less than that when mass ratio is 50%. However, active component surface is covered when the mass ratio is over-high, which causes the rapid decrease of basic sites that is crucial in the transesterification reaction. Thus, the ratio of 50% is thought as the best mass ratio of K_2CO_3 to HT.

In Fig. 6, the impact of molar ratio of methanol to oil on FAMEs yield is shown. FAMEs yield increased from 40.67% to 96.69% when the molar ratio increased from 6:1 to 12:1. But, the yield reduced gradually when the molar ratio increasing from 12:1 to 18:1. Concentrations change of curcas oil and methanol was thought as a main reason for this phenomenon. From the molar ratio being 6:1 to 9:1, oil concentration was relatively high in

Fig. 5 Effect of mass ratio of K_2CO_3 to HT on yield of FAMEs. Reaction condition: temperature was 65 °C, molar ratio was 12:1, catalyst amount was 3%, and reaction time was 3 h



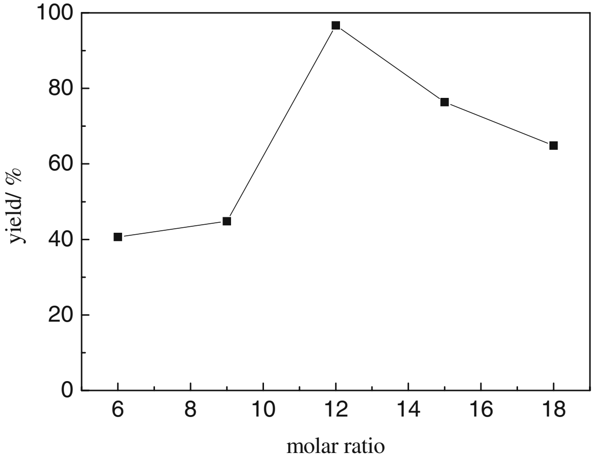


Fig. 6 Effect of molar ratio of methanol to oil on yield of FAMEs. Reaction condition: catalyst amount was 3%, reaction time was 3 h, mass ratio was 50%, and reaction temperature was 65 °C

reactant mixture liquid, while methanol concentration was very low. Little methanol supplied fewer molecules, general collision times were less therefore. For this reason, yield of FAMEs was low when the molar ratio of methanol to oil was lower than 12:1. On the contrary, the concentration of oil became low when the molar ratio was 15:1 and 18:1, in which overall collision times became less under the same reaction conditions. With the ratio increasing, less crash times lowered the yield of FAMEs. Therefore, 12:1 was accepted as the suitable and appropriate molar ratio for the transesterification reaction.

Catalyst amount was another crucial influence in this reaction. When molecules of curcas oil and methanol crashed on the active sites, transesterification reactions happened. Therefore, quantity of basic active sites on catalyst surface was pivotal to reaction. Effect of catalyst amount on yield of FAMEs was shown in Fig. 7. The yield of FAMEs kept rising along with the catalyst increasing. The more catalyst amount was provided, the more basic active sites was gained, which could be a good explanation to the dramatic increase of FAMEs yield. However, basic sites were nearly enough for the transesterification reaction when the catalyst was 3% of curcas oil (by weight). In spite of

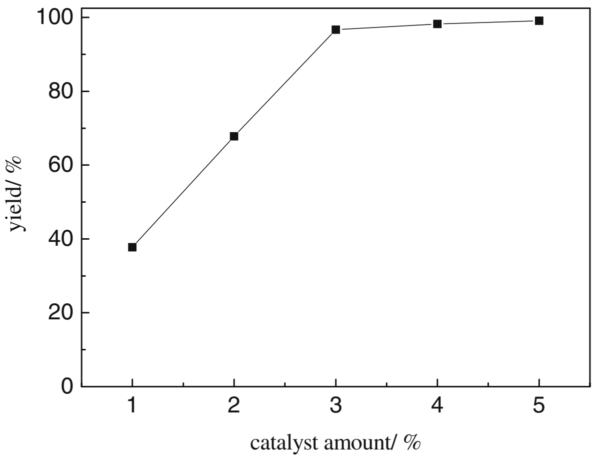


Fig. 7 Effect of catalyst amount on yield of FAMEs. Reaction condition: reaction time was 3 h, mass ratio was 50%, reaction temperature was 65 °C, and molar ratio was 12:1

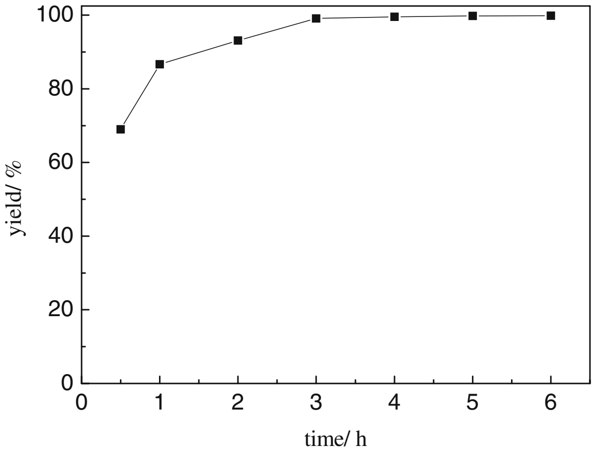


Fig. 8 Effect of reaction time of K_2CO_3 to HT on yield of FAMEs. Reaction condition: mass ratio was 50%, reaction temperature was 65 °C, molar ratio was 12:1, and catalyst amount was 3%

addition amount of catalyst, the yield could not increase sharply when amount was over 3% (by weight). Therefore, 3% of oil was accepted as the optimal catalyst amount where the yield was 96.69%.

As another important influence factor in the transesterification reaction, reaction time was investigated, which was showed in Fig. 8. Longer reaction time meant more contact time between curcas oil and methanol molecules on basic sites of catalyst. Likewise, more contact time brought more chance for the reaction and then, more molecular methanolysis reactions between triglycerides and methanol occurred. Thus, it was a good explanation why the yield of FAMEs was increasing with the reaction occurring. However, owing to the equilibrium of the reaction, the increase of yield was nearly halted when the time was more than 3 h. After the reaction had lasted for 3 h, reaction equilibrium was almost reached which also meant plus and minus reaction speed was equal. It was thought as a good explanation why the yield increased slightly from reaction time being 3 to 6 h. That is why 3 h was selected as the best reaction time.

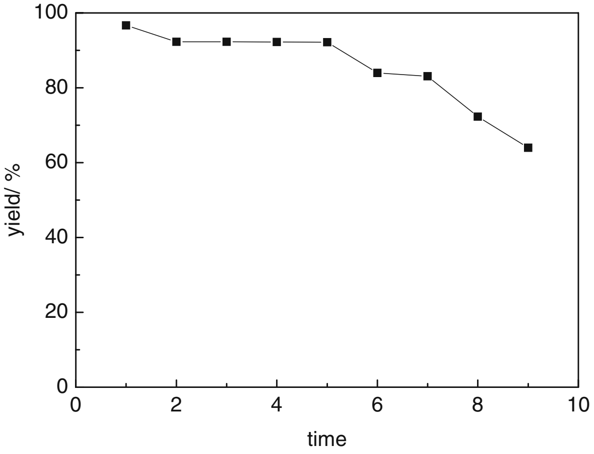


Fig. 9 Effect of repeated times on yield of FAMEs. Reaction condition: mass ratio was 50%, reaction temperature was 65 °C, molar ratio was 12:1, and catalyst amount was 3 %

Table 2 Yields of biodiesel from *Jatropha curcas* oil using different catalysts.

Optimum conditions	*Jatropha curcas* oil+methanol			
Catalyst	KNO_3/Al_2O_3	CaO	NaOH	K_2O_3/HT
Load ratio	35%	–	–	50%
Molar ratio	1:12	1:9	MeOH to Oil=70% (by weight)	1:12
Temperature	70 °C	70 °C	65 °C	65 °C
Catalyst mass (by weight of oil)	6 %	1.5 %	3.3 %	3 %
Reaction time	6 h	2.5 h	2 h	3 h
Yield	84%	93%	55%	96.69%

Data from Zhu et al., Vyas et al., and Berchmans et al. [31, 32, 41]

Repeated Experiments

Lifespan of catalyst was of much importance for its practical application and economical effect. Thus, the repeated experiments were finished. Effect of repeated time of catalyst on biodiesel yield was showed in Fig. 9. With the usage times going up, the yield of biodiesel decreased gradually. However, in the first five times, the yields of biodiesel could keep more than 92%. Therefore, it is considered that the catalysis effect was kept stable during the first five times. It also proved that the morphology changed very slightly after being used five times.

Comparisons

In order to show the catalysis effect, comparison among some other basic catalysts were carried out, which was illustrated in Table 2 below. In all of these experiments, *Jatropha curcas* oil and methanol were used as raw materials. Compared with the other results, yield of biodiesel using K_2O_3/HT as a catalyst was the highest. What is more, the reaction conditions were relatively mild.

In order to test whether characteristics of the product could meet ASTM standard, product properties comparison with ASTM was carried out. Table 3 listed some parameters and corresponding ASTM standard. As for cloud point, it is a little higher than the standard.

Table 3 Comparison of the product properties with ASTM standard.

Parameters/unit	This product	ASTM D6751-07a standard
Flash point/°C	127	93
Water content/ %	0.03	0.05
Viscosity, 40 °C/ mm^2/s	3.27	1.9–6.0
Cetane number/	49	47
Free glycerol/ %	0.19	0.2
Total glycerol/ %	0.20	0.24
Acid number/ mg KOH/g	0.43	0.50
Cloud point/°C	−12	−19
Sulfur content/ %	0.00015	0.05

ASTM D6751, 2007

However, as seen in Table 3, all the listed parameters could meet ASTM standard, except the cloud point. Therefore, the properties of this product were considered acceptable.

Conclusion

In this paper, the preparation of biodiesel from *Jatropha curcas* oil was discussed. In the experiments, the optimal transesterification reaction conditions were as follows: the mass ratio of K_2CO_3 to HT was 50%, the molar ratio of methanol to oil was 12:1, the reaction temperature was 65 °C, the catalyst amount was 3% of oil (by weight), and the reaction time was 3 h. Under the above conditions, the yield of biodiesel could reach up to 96.69%. According to comparison with ASTM standard, this product could meet the standard.

Acknowledgment The authors are grateful to National High Technology Research and Development Program of China (No.2009AA03Z222 and No.2009AA05Z437) and “Six Talents Pinnacle Program” (No.2008028) of Jiangsu Province of China for financial support.

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