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Calcium-based mixed oxide catalysts for methanolysis of *Jatropha curcas* oil to biodiesel

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ABSTRACT

Calcium-based mixed oxides catalysts (CaMgO and CaZnO) have been investigated for the transesterification of *Jatropha curcas* oil (JCO) with methanol, in order to evaluate their potential as heterogeneous catalysts for biodiesel production. Both CaMgO and CaZnO catalysts were prepared by coprecipitation method of the corresponding mixed metal nitrate solution in the presence of a soluble carbonate salt at ~ pH 8–9. The catalysts were characterized by X-ray diffraction (XRD), temperature programmed desorption of CO_2 (CO_2-TPD), scanning electron microscopy (SEM) and N_2 adsorption (BET). The conversion of JCO by CaMgO and CaZnO were studied and compared with calcium oxide (CaO), magnesium oxide (MgO) and zinc oxide (ZnO) catalysts. Both CaMgO and CaZnO catalysts showed high activity as CaO and were easily separated from the product. CaMgO was found more active than CaZnO in the transesterification of JCO with methanol. Under the suitable transesterification conditions at 338 K (catalyst amount = 4 wt. %, methanol/oil molar ratio = 15, reaction time = 6 h), the JCO conversion of more than 80% can be achieved over CaMgO and CaZnO catalysts. Even though CaO gave the highest activity, the conversion of JCO decreased significantly after reused for forth run whereas the conversion was only slightly lowered for CaMgO and CaZnO after sixth run.

1. Introduction

With concern on the availability of recoverable petroleum reserves and the impact of environmental pollution of exhaust gases from fossil-based fuels, biodiesel has received more and more attention as a promising alternative fuel [1,2]. Biodiesel, a mixture of fatty acid methyl esters (FAMES) which possesses similar physicochemical and fuel properties to the petroleum based diesel fuel. Biodiesel is a clean-burning fuel derived from vegetable oils or animal fat and is an advantageous alternative to fossil diesel fuel because of its biodegradability, biorenewable nature, low sulfur content and toxicity, low volatility/flammability, good transport and storage properties, higher cetane number, and its salutary atmospheric CO_2 balance for production [3–5]. Treatments that have been done to avoid the significant problems associated with the high viscosity of plant oils when used as engine fuels are its dilution, microemulsification, pyrolysis and transesterification. Transesterification with methanol is most widely used for this purpose [6]. The glycerol by-product formed during the

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transesterification can be used in numerous applications in the food, cosmetic, and pharmaceutical sectors [7].

In fact, biodiesel has already been commercially produced and entered the fuel market as diesel substitute. It is normally manufactured by transesterification of triglyceride with alcohol which is also known as alcoholysis, in which glycerol in oil is displaced by low chain alcohols [7].

At present day, the cost of biodiesel is not competitive to petroleum diesel without subsidies or tax incentives as the cost of raw materials and processing are high. The cost of crop oil account for a large percent in direct biodiesel production cost which includes capital cost and returns [8]. Besides, the availability of oil crop for the biodiesel production is limited [9]. Therefore, it is necessary to find new feedstock suitable for biodiesel production, which does not drain on the edible vegetable oil supply. With no competing food uses, this characteristic turns attention to *Jatropha curcas*, which grows in tropical and subtropical climates across the developing world [10].

In Malaysia, wild *Jatropha curcas* tree is also known as "jarak pagar" particularly in Peninsular Malaysia area. *Jatropha curcas* oil can not be used for nutritional purposes without detoxification makes its use as an alternative or greener oil resource for biodiesel synthesis. *Jatropha curcas* (Linnaeus) is a multipurpose bush/small tree belonging to the Euphorbiaceae family. The plant can be planted as a hedge to prevent field's erosion, to reclaim land and grown as a live fence, especially to contain or exclude farm animals and also planted as a commercial crop. *Jatropha curcas* can grow well under adverse climatic because of its low moisture demands, low fertility requirements and tolerance to high temperatures. It is widely cultivated in central and south America, south east Asia, India and Africa [11].

Commercial transesterification processes usually uses H_2SO_4, HCl, NaOH, or KOH as catalysts. The homogeneous catalysts often result in many problems such as equipment corrosion and some environmental problems from the deposit of waste catalysts [12]. The cost of biodiesel could certainly be reduced by using heterogeneous catalyst. The reusability and ecofriendly nature of heterogeneous catalyst lend themselves to easier product separation and better product purity since water washes can be eliminated. In contrast, processes using heterogeneous catalysts require higher temperatures to be effective in a reasonable time [13].

Different solid catalysts for biodiesel production have been proposed, mainly based on basic alkaline and alkaline-earth compounds [14,15], acid solids [16,17] and enzymes [18]. Moreover, the transesterification of vegetable oils also performed in supercritical methanol without catalyst [19]. The transesterification catalyzed by solid base catalysts show good catalytic activity under the mild reacting condition as compared to solid acid catalysts [20]. However, most of these catalysts probably catalyzed transesterification through a homogeneous molecular pathway rather than a truly heterogeneous process due to their non-negligible solubility in alcohols [21]. Nevertheless, studies on the reusability and regeneration of the catalysts are necessary to estimate the potential of a specific catalyst for commercial application.

Recently, much interest has been taken in calcium oxide (CaO) due to its economical advantages. Kawashima and coworkers [22] found that calcium oxide was so active (90% of biodiesel yield) as to finish transesterification of rapeseed oil with refluxing methanol within a reaction time of 3 h. Reddy et al. [13] reported that high catalytic activity of nanocrystalline CaO (99% conversion) in the transesterification reaction of poultry fat and soybean oil at room temperature. Also, Liu et al. [23] investigated the effect of moisture on the catalytic activity of CaO. However, some researchers have pointed out that soluble substance leached away from CaO during the transesterification. Gryglewicz reported that CaO was slightly dissolves in methanol and form suspension in the reaction mixture due to its poor mechanical strength. The suspension will result in separation problems of catalyst from biodiesel products [14]. Therefore, supporting CaO onto carriers may suggest improving the stability of CaO catalysts.

The present work investigated the behaviours of several binary oxides of calcium-based solid catalysts (CaMg and CaZn oxides) in methanolysis of *Jatropha curcas* oil for biodiesel production. All precursors were synthesized by using the coprecipitation method, and after calcination the corresponding metal oxides were obtained. The structural, textural and basic characteristics of catalysts were determined by different techniques.

2. Experimental section

2.1. *Catalysts preparation*

The mixed metal oxide between calcium with magnesium (CaMgO) and calcium with zinc (CaZnO) were prepared by the coprecipitation method, and follows calcination of the precursors. In a typical method of catalysts preparation, 2 M aqueous solution of the corresponding metal nitrates ($Ca(NO_3)_2.4H_2O$, $Mg(NO_3)_2.6H_2O$ and $Zn(NO_3)_2.6H_2O$), were slowly added into basic aqueous solution that containing Na_2CO_3 and NaOH at a constant pH of 8–9. The resulting precipitations were performed under vigorous stirring at 338 K for overnight. Finally, the solids were filtered and washed with deionized water. The resultant precursors were dried in oven overnight. These precursors were calcined at 1073 K and 1173 K for 6 h in which the former denoted as CaMgO and later CaZnO, respectively.

2.2. *Characterization*

The powder X-ray diffraction analysis was carried out using a Shimadzu diffractometer model XRD6000. The diffractometer employing CuK_α radiation to generate diffraction patterns from powder crystalline samples at ambient temperature. The $Cu\text{-}K_a$ radiation was generated by Philips glass diffraction X-ray tube broad focus 2.7 kW type. All samples were mounted on samples holder and the basal spacing was determined via powder technique.

The total surface area of the catalysts was obtained by Brunauer-Emmer-Teller (BET) method using nitrogen adsorption at 77 K. The analysis was conducted using Thermo Finnigan Sorptomatic 1900 series nitrogen adsorption/desorption analyzer.

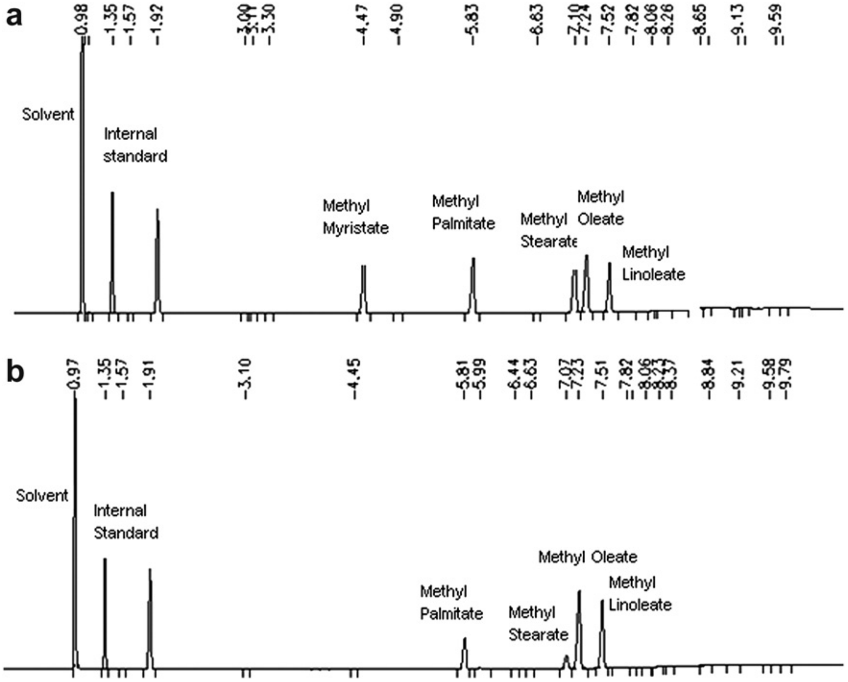

Fig. 1 – Gas chromatogram of (a) standard methyl ester and (b) experimental biodiesel product.

The scanning electron microscopy (SEM) technique was used to obtain the information of the morphology and size of the samples. The morphology of the catalysts was carried out using a Hitachi s-3400 n electron microscope. The samples were coated with gold using a Sputter Coater.

The basicity of the catalysts were studied by temperature programmed desorption using CO_2 as probe molecule (TPD-CO_2). TPD- CO_2 experiment was performed using a Thermo Finnigan TPDRO 1100 apparatus equipped with a thermal conductivity detector. Catalysts (100 mg) were pretreated under a helium stream at 1073 K for 30 min (10 K min^{-1}, 20 mL min^{-1}). Then, the temperature was decreased to 303 K, and a flow of pure CO_2 (20 mL min^{-1}) was subsequently introduced into the reactor for 1 h. The sample was flushed with helium at 303 K for 30 min before the CO_2 desorption analysis. The analysis of CO_2 desorption was then carried out between 373 and 1173 K under helium flow (10 K min^{-1}, 20 mL min^{-1}) and detected by thermal conductivity detector (TCD).

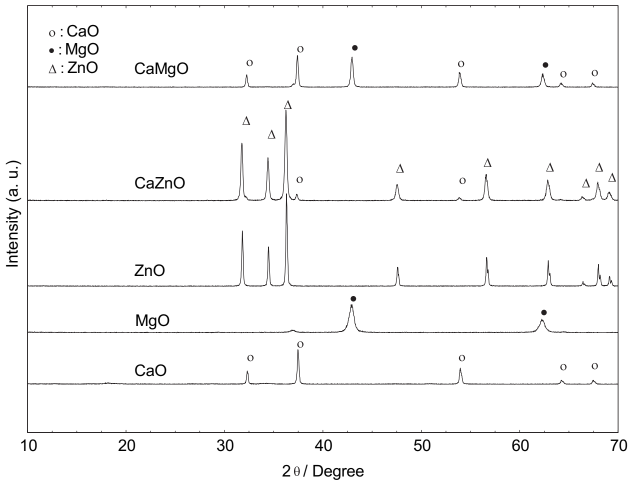

Fig. 2 – XRD patterns of CaO, MgO, ZnO, CaMgO and CaZnO catalysts.

The bulk chemical composition of catalysts was determined by using inductively coupled plasma-atomic emission spectrometer (ICP-AES) analysis that conducted by using Perkin Elmer Emission Spectrometer Model Plasma 1000.

2.3. *Transesterification procedure*

Crude *Jatropha curcas* oil (JCO) was purchased from Bionas Sdn. Bhd. Methanol was commercial grade with 99.5% purity. These liquids were used without further purification. Transesterification was carried out in a 150-mL 2-neck round bottom flask equipped with a reflux condenser and a magnetic stirrer. Typically, CaMgO and CaZnO (1–10 wt. %) were suspended in a required volume of methanol. The temperature of the mixture was controlled at 338 K by a water bath. Subsequently, oil was added into the mixture under vigorous stirring. The methanol/oil molar ratio was varied between 5 and 25. After the course of reaction (2–12 h), the catalyst was separated via centrifugation and the reaction mixture was then loaded into a rotary evaporator to remove excess methanol. Experiments were carried out by changing different parameters like methanol/oil molar ratio, reaction time and catalyst amount. Conversion of JCO to biodiesel by CaMgO and CaZnO were compared with pure metal oxide (CaO, MgO and ZnO) catalysts.

Since the transesterification of triglycerides can produce methyl ester (biodiesel) and by-product, glycerol. Hence, the conversion of oil was calculated based on the amount of recovered glycerol. Glycerol formed after the reaction was weighted and moles of the glycerol were calculated. This mole value of glycerol is compared with theoretical value and based

Table 1 – Bulk chemical composition analysis and physicochemical properties of CaO, MgO, ZnO, CaMgO and CaZnO catalysts.

Catalysts	Theoretical molar ratio[a]	Experimental molar ratio[a]	Crystallite size (nm)[b]			S_{BET} (m^2g^{-1})[c]
			CaO	MgO	ZnO	
CaO	–	–	40.2	–	–	9.5
MgO	–	–	–	14.4	–	29.7
ZnO	–	–	–	–	58.8	4.9
CaMgO	0.25	0.30	43.8	35.6	–	15.5
CaZnO	0.25	0.22	41.6	–	36.7	9.8

a Estimated from ICP–AES analysis.
b Crystallite sizes were calculated accordingly to Scherrer's equation [26].
c BET surface area.

on this difference conversion of triglyceride was found out [24,25].

$$\text{Conversion}(\%) = \frac{\text{Mole of experimental glycerol}}{\text{Mole of theoretical glycerol}} \times 100\% \quad (1)$$

The biodiesel product was analyzed by gas chromatography using Perkin Elmer with FID detector, equipped with a HP INNOwax capillary column. Dichloromethane was used as an internal standard. The gas chromatogram of the biodiesel product was shown in Fig. 1.

3. Results and discussion

3.1. *Characterization of materials*

The XRD patterns of catalysts (CaO, MgO, ZnO, CaMgO and CaZnO) were shown in Fig. 2. XRD diffractogram of CaO gave intensified and narrowed peaks at 2θ of 32.2°, 37.3° and 53.8° (JCPDS File No. 37-1497). For MgO, the diffraction pattern clearly observed at 2θ angles of 42.9° and 62.2° (JCPDS File No. 4-0829). The XRD pattern of ZnO showed, main crystallite phases at 31.8°, 34.5° and 36.3° at 2θ angles (JCPDS File No. 36-1451). In the case of CaMgO, the thermal treatment gives rise to the formation of cubic CaO ($2\theta = 32.2°$, 37.3° and 53.8°) and hexagonal MgO ($2\theta = 42.9°$ and 62.3°). Whereas, The CaZnO catalyst showed diffraction peaks of CaO at 2θ angles of 37.4° and 53.9° and ZnO at 2θ angles of 31.7°, 34.4° and 36.2° with wurzite structure. These results indicated that the crystalline phases present in both mixed oxides catalysts (CaMgO and CaZnO) were corresponded to pure oxide, there was no new crystalline phase of mixed oxides could be detected, this was mainly due to the different ionic radii of the metal ions.

Chemical analysis, crystallite size and surface area of the catalysts were summarized in Table 1. The result revealed that the Ca/Mg and Ca/Zn molar ratio were close to the theoretical values with 0.30 and 0.22, respectively. The crystallite size of catalysts can be calculate from the line broadening or FWHM (full width at half maximum) of corresponding peaks (the most intense peaks), which is by using Debye-Scherrer's equation [26]:

$$t = \frac{0.89\lambda}{\beta_{hkl}\cos\theta_{hkl}} \quad (2)$$

Where t is the crystallite size for (hkl) phase (nm), λ is the X-ray wavelength of radiation for CuKα, β_{hkl} is the full width at half maximum (FWHM) at (hkl) peak in radian and θ is the diffraction angle for (hkl) phase. In this case, the average crystallite sizes for pure CaO, MgO and ZnO were 40.2, 14.4 and 58.8 nm, respectively. The binary oxides catalysts (CaMgO and CaZnO) showed the metal oxides clusters with 43.8 nm and 41.6 nm in crystallite size for CaO, 35.6 nm for MgO and 36.7 nm for ZnO. The results indicated that crystal sizes of catalysts were in agreement with the line width of the peak in which decrease of FWHM with the increment of the crystallite size. The surface area of CaO, MgO, ZnO, CaMgO and CaZnO were shown in Table 1. It was found that the catalysts with surface area were arranged in the sequence of: MgO > CaMgO > CaZnO > CaO > ZnO. The surface area of the catalysts was in agreement with XRD analysis which showed that larger crystallite sizes gave lower surface area [27].

The total basicity and basic site distributions were evaluated by using temperature programmed desorption of CO_2. The CO_2-TPD profiles and amount of basicity of the catalysts were displayed in Fig. 3 and Table 2, respectively. The TPD patterns of the catalysts showed CO_2 desorption peaks at the temperature ranging from 400 to 700 K and 800–1000 K. According to Di Serio et al., a very broad desorption band extending from 400 to 700 K could be assigned as interaction of CO_2 with sites of weak and medium basic strengths. The narrower band at higher temperature (between 700 and 900 K)

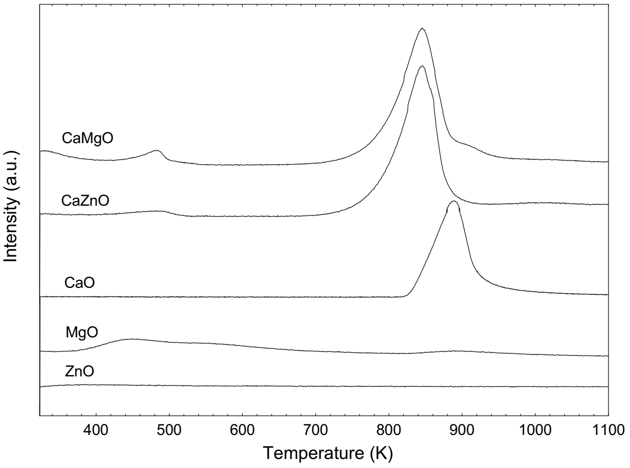

Fig. 3 – CO_2-Temperature programmed desorption profiles of CaO, MgO, ZnO, CaMgO and CaZnO catalysts.

Table 2 – The basic intensity of CaO, MgO, ZnO, CaZnO and CaMgO catalysts.

Catalysts	CO_2 desorbed (μmol/g)	Temperature range (K)	Peak temperature (K)
CaO	290.42	820–1021	888
MgO	31.36	374–720	449
	18.95	839–972	895
ZnO	–	–	–
CaZnO	412.7	793–1000	902
CaMgO	2.68	470–526	505
	449.7	757–912	859

can be attributed to the presence of much stronger basic site [28]. The CaO gave strong basic strength with an intense desorption peak at 888 K, whereas the pure MgO showed a broad desorption peak with mainly medium basic site at 449 K and a small weak peak with strong basic strength at 895 K. However, no CO_2 desorption peak was observed in the TPD spectra for ZnO catalyst. This showed that there is no basic site on the surface of ZnO. As for CaMgO, the first desorption peak occurred at 484 K can be ascribed to the presence of MgO with weak and moderate basic sites, whereas the peak located at 846 K indicated the existence of strong basic sites resulted from introduction of Ca^{2+} to the binary oxide. On the other hand, CaZnO showed the presence of one intense peak at 847 K with higher CO_2 desorption volume as compared to other catalysts. In the Ca-Zn binary system, the strong basic sites was generated as ZnO plays an important role by giving strong interaction between CaO and ZnO leading to the transfer of electrons from metal zinc oxide support interface to CaO [29]. The TPD results indicated that mixed metal oxides systems provide stronger basic strength on the catalytic site of catalysts as compared to pure metal oxides.

The morphology of the catalysts was studied by scanning electron microscopy (SEM). SEM images of CaO, MgO, ZnO,

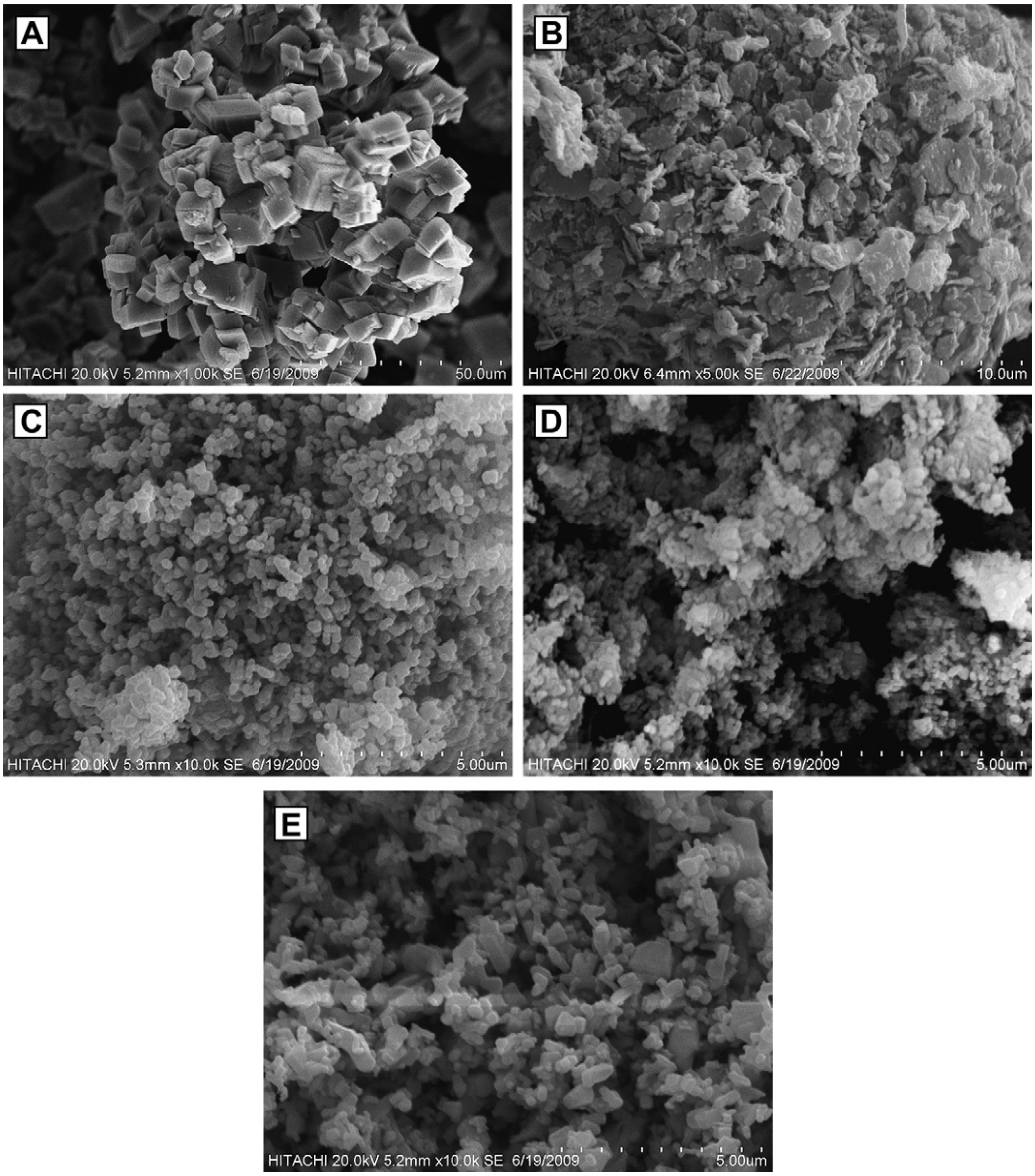

Fig. 4 – SEM images of CaO(A), MgO(B), ZnO(C), CaMgO(D) and CaZnO(E) catalysts.

CaMgO and CaZnO were shown in Fig. 4. The SEM image of CaO showed a cluster of well-developed cubic crystal with obvious edges (Fig. 4A). SEM micrographs of MgO gave plate like structure with hexagonal shape (Fig. 4B) and ZnO with small round-shape particles on its surface (Fig. 4C). Both CaMgO and CaZnO (Fig. 4D and E) gave tiny aggregates particles on the surface of catalysts. It can be observed clearly from SEM images that the morphology for CaMgO was totally different from the pure CaO and MgO. The CaMgO sample with compact agglomeration was in agreement with the morphology that has been noted previously for MgO/CaO which is dense particles with a heterogeneous distribution of particle sizes [30]. In the case of CaZnO, the image showed the homogeneous form of small spherical particles which have similar structure as pure ZnO. The homogeneous distribution of ZnO particle in CaZnO was in agreement with the results from XRD and ICP-AES analysis in which the CaO content in the catalyst was low.

3.2. *Catalytic activity in transesterification process*

The transesterification catalytic activity of CaO, MgO, ZnO, CaMgO and CaZnO catalysts were indicated in Table 3. The reactions were carried out at transesterification conditions: catalyst amount, 4 wt. %; methanol/oil ratio, 15:1; reaction temperature, 338 K and reaction time, 6 h. Among the catalysts, CaO showed the highest catalytic activity with 85% JCO conversion. The conversion of JCO was found very low when using MgO catalyst (~10%) and non active ZnO did not produced biodiesel in the reaction. Interestingly, calcium-based mixed metal oxides (CaMgO and CaZnO) showed encouraging result in converting JCO to biodiesel with 83% and 81%, respectively.

3.3. *Effect of preparation parameters on JCO conversion*

3.3.1. *Catalyst amount*

The effect of catalysts (CaO, CaMgO and CaZnO) dosages on the conversion of JCO was examined. The catalyst amount was varied in the range of 1.0% and 10.0% with referred to the starting oil weight. From the reaction profile of Fig. 5, the conversion of JCO using CaO catalyst was increased gradually (over 80%) with the increment of catalyst amount from 1 to 6 wt. %. The conversion started to decrease with further addition of CaO amount. This result is probably due to resistant of mixing involving reactant, product and solid catalyst. Besides, more biodiesel products were adsorbed into the catalyst as the dosage of CaO was too high, and this reduced the biodiesel content. The calcium-based mixed metal oxides (CaMgO and CaZnO) showed gradual increment of conversion as the amount of catalysts is being increased.

Table 3 – Conversion of *Jatropha curcas* oil (JCO) (%) from transesterification reaction by CaO, MgO, ZnO, CaMgO and CaZnO catalysts.

Catalysts	Conversion of JCO (%)
CaO	85
MgO	10
ZnO	–
CaMgO	83
CaZnO	81

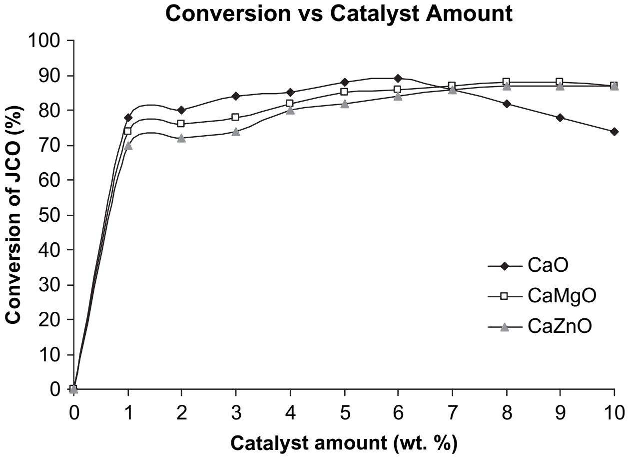

Fig. 5 – Influence of catalysts amount (CaO, CaMgO and CaZnO catalysts) on the conversion of JCO. Reaction condition: Jatropha oil 10 g, reaction time 6 h and reaction temperature 338 K, methanol/oil molar ratio 15:1.

3.3.2. *Reaction time*

The conversion of JCO versus reaction time was presented in Fig. 6. The presence of heterogeneous mass transfer systems led to the slow reaction within short reaction time. Hence, it is assumed that the conversion was low in the first 1 h. However, it can be seen that the conversion for these three catalysts were increased gradually after 2 h of reaction time, and thereafter remained nearly constant. The nearly equilibrium conversion was found to be around 80% at 6 h of reaction time. The CaO catalyst showed the highest conversion in shorter time as compared to CaMgO and CaZnO, this was due to the slightly solubility of CaO catalyst in the liquid reaction.

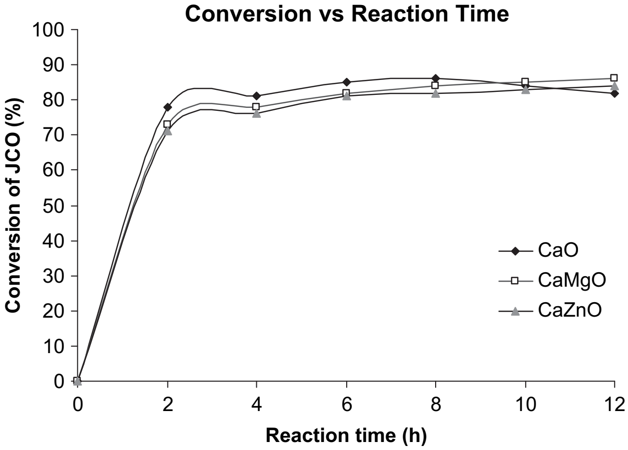

Fig. 6 – Influence of reaction time on the conversion of JCO by CaO, CaMgO and CaZnO catalysts. Reaction condition: Jatropha oil 10 g, catalyst amount 4 wt. %, reaction temperature 338 K and methanol/oil molar ratio 15:1.

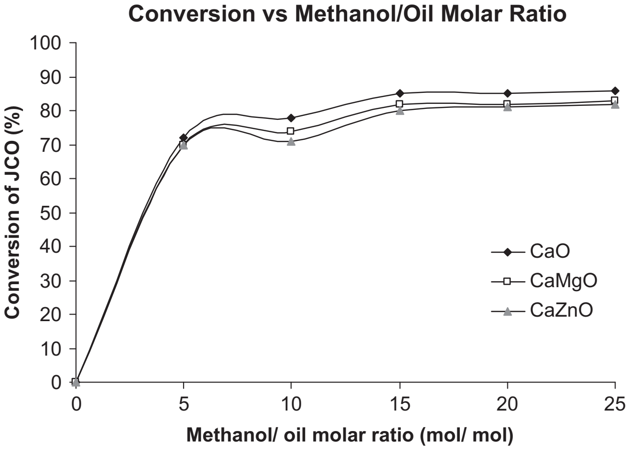

Fig. 7 – Influence of methanol/oil ratio on the conversion of JCO by CaO, CaMgO and CaZnO catalysts. Reaction condition: Jatropha oil 10 g, catalyst amount 4 wt. %, reaction temperature 338 K and reaction time 6 h.

However, the appearance of white emulsion and gel in the product was observed which resulted from further prolongation of reaction time. This increased the viscosity of the product and might influence the purification process. The conversion of JCO by CaMgO and CaZnO increased gradually with the increment of reaction time.

3.3.3. *Methanol/oil molar ratio*

Stoichiometrically, one mole of triglyceride requires three moles of methanol for the transesterification reaction in order to produce fatty acid methyl ester or biodiesel. Triglycerides transesterification is a reversible reaction, excess of methanol is used to drive the reaction towards completion of ester formation. However, excessive methanol is not favorable for the purification of the as-synthesized biodiesel as more energy is needed to recover the excessive of methanol. Besides, excess methanol can increase the dissolution of oil, intermediates and biodiesel product which resulting in the wastage of the raw reactants. According to the result from Fig. 7, the conversion of JCO with the presence of the catalysts exhibited a strong dependence on the methanol to oil ratio. When the methanol loading amount was increased, the conversion was increased considerably. The maximum conversion was obtained at a molar ratio of approximately 15:1, beyond which additional methanol had little improvement in the conversion.

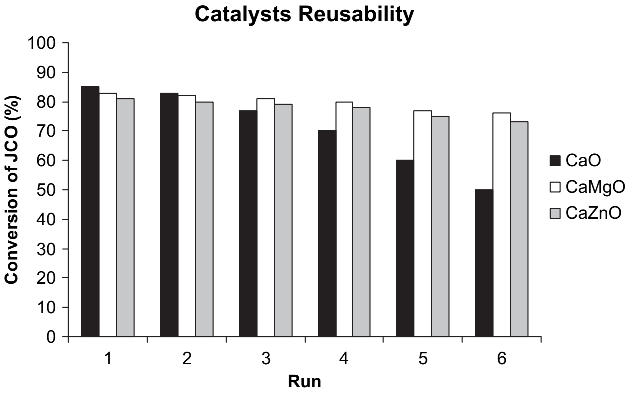

Fig. 8 – Reusability study of CaO, CaMgO and CaZnO catalysts. Reaction condition: Jatropha oil 10 g, catalyst amount 4 wt. %, reaction temperature 338 K, reaction time 6 h and methanol/oil ratio 15:1.

Table 4 – ICP-AES analysis of fresh and used CaO, CaMgO and CaZnO.

Catalyst	CaO	CaMgO		CaZnO	
Concentration of element (ppm)	Ca	Ca	Mg	Ca	Zn
Fresh catalyst	44.2	17.9	36.2	10.0	74.3
Fifth used catalyst	9.0	12.8	28.5	6.3	62.8

3.4. *Catalyst reusability*

One of the most important features for a catalyst to be industrially useful is its stability and reusability. Some recent studies [14] showed a fraction of bulk CaO is dissolved in the methanolic solution, and leaching from catalyst will eventually reduce the activity of catalyst. Hence the objective of preparing mixed calcium-based oxides (CaMgO and CaZnO) was intended to stabilize the active phase in order to avoid the leaching process. The three catalysts were recycled to study the reusability. The used catalysts were washed with hexane and methanol to removed organic deposit (unreacted oil, biodiesel and glycerol). The CaMgO and CaZnO were then dried in oven for overnight and calcined for 3 h at 1073 K and 1173 K, respectively, in order to burn off the entire unwanted organic deposit product and regenerated the active sites of the catalysts. After calcination, catalysts were used for next run. The data of reusability of the catalysts were presented in Fig. 8, it showed that the conversion of JCO by CaO catalyst decrease continuously for five runs. However, the conversion of JCO catalyzed by CaMgO and CaZnO catalysts decreased slightly for next five runs. These results suggested that the loss of transesterification activity in the present case mainly derived from the dissolution of active metal and deposition of organic materials on the active surface. CaMgO and CaZnO catalysts can be repeatedly used up to 4 times while maintaining the biodiesel yield at around 80% in the transesterification reaction.

The ICP-AES analysis was performed to study the leaching of calcium active phases for CaMgO and CaZnO catalysts. The result for the fresh and fifth used catalyst was depicted in Table 4. It is showed that the calcium active content drastically dropped for CaO but slightly reduced for calcium-based mixed oxide catalysts. As a result, the transesterification activity of CaMgO and CaZnO able to retain for fourth run but the activity of CaO was remarkably decreased.

4. Conclusions

In the present study, CaMgO and CaZnO mixed oxides have been synthesized and their catalytic performances in the transesterification of *Jatropha curcas* oil have been investigated and compared with reference catalysts (CaO, MgO and ZnO). CaMgO and CaZnO were active as basic catalysts for transesterification processes, which could be attributed to the presence of strong basic sites on the surface, mainly associated to Ca^{2+}-O^{2-} pairs. The surface area of CaMgO and CaZnO were much higher than that of CaO and ZnO but lower than MgO due to the different of crystallite sizes. By varying the transesterification conditions at 338 K, more than 80% of JCO conversion achieved when the reaction parameters were set as follows: the methanol/oil ratio = 15, the amount of CaMgO and CaZnO catalysts = 4 wt. % (based on weight of the oil), and the reaction time = 6 h. The CaZnO and CaMgO catalysts can be reused up to 3 and 4 times, respectively with maintaining the conversion>80%.

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