Effect of MgO Loading on the Production of Biodiesel from Jatropha Oil in the Presence of MgO/MCM-22 Catalyst

Bashir Abubakar ABDULKADIR※1, Anita RAMLI ※1†, Lim Jun WEI※1, and Yoshimitsu UEMURA ※2

(Received November 21, 2017)

This research focuses on the preparation, characterization and catalytic properties of novel magnesium oxide in mesoporous MCM-22 Silica (MgO/MCM-22) for biodiesel production from jatropha oil. The catalysts were prepared by impregnation method and characterized. Then, the catalyst were tested for biodiesel production from jatropha oil where the reaction parameters were studied. The obtained results of the characterization showed that, the prepared catalyst has an amorphous structure with high Si/Al ratio which changes to a more of crystalline structure after successful incorporation of MgO. The result of the transesterification shows that, magnesium oxide (MgO) supported with silica is highly effective for the production of biodiesel where the high fatty acid methyl ester of 98% was achieved. Furthermore, the synthesized catalyst was able to be used up to three times with a slight reduction in catalytic activity. Hence, it can be concluded that the catalyst prepared from MgO/MCM-22 can serve as an outstanding alternative catalyst for biodiesel production.

Key Words
Transesterification, MgO, MCM

1. Introduction

One of the important goals of catalysis research is to synthesize and design a catalyst that can produce only one preferred product out of many other possible side products, that is, 100% selectivity 1). Such green technology of heterogeneous catalyst will help in eliminating the production of any unwanted products (wastes). To synthesize such type of catalyst for the green technology, consideration of the molecular ingredients that influence selectivity must be incorporated into the catalyst by incorporating metal oxide into a catalyst support. It was reported that, heterogeneous catalysts are non-corrosive, are environmentally friendly, can be easily separated from the products via filtration, and present fewer disposal problems than those encountered by the homogeneous catalysts 1). In this study, bifunctional heterogeneous catalyst was prepared by a modified impregnation method and characterized by various analytical techniques. The synthesized catalysts were tried in the transesterification reaction of jatropha oil for low cost biodiesel production where the best catalyst with metal (MgO) loading that give higher FAME yield was obtained. Hence, this research aimed to prepare, synthesize and characterize low cost, highly effective MgO in MCM-22 supported catalyst for transesterification reaction.

2. Experimental

2.1 Material and Methods

The MCM-22 and the magnesium nitrate (Mg(NO₃)₂·6H₂O) used in this study were purchased and supplied by ACS Materials, LLC Advanced Chemical Supplier and it was used as received without any modification. The zeolite was calcined in a muffle furnace at a temperature of 500 °C for 3 hours. Jatropha oil was purchased from Bionas Green and Clean Energy Products, Malaysia. The obtained oil was then dried in an oven at 80 °C where all the remaining moisture was evaporated and subsequently filtered where all the unwanted particles that might stick to the oil were removed.
2.2 Catalyst preparation

The catalyst containing different loadings of MgO with respect to MCM-22 silica were prepared by wet impregnation method as describe in the work of Farooq et al. The required quantity of magnesium nitrate hexahydrate (Mg(NO₃)₂.6H₂O) supplied by Merck with 99.99% purity was dissolved in a beaker containing distilled water and strongly stirred. Subsequently, the required amount of already calcined powdered MCM-22 was carefully added into the beaker containing the solution of magnesium nitrate hexahydrate and stirred. The mixture was stirred constantly for about 3 h at a temperature of 80 °C followed by drying in an oven at 110 °C for 24 h. Finally, the obtained solid sample was calcined at temperature of 500 °C for 3 h in a muffle furnace to get MgO/MCM-22 with different MgO loadings.

2.3 Catalyst characterization

The crystalline phases of the prepared catalysts were analyzed by X-ray diffraction and it was conducted using Bruker D8 Advance Power diffractometer with Cu Kα radiation (λ=1.54Å) for a 2θ scattering angle in the range of 20 ° to 100 ° at scanning rate 5 °/min. Field-Emission Scanning electron microscopy/EDX (FESEM-EDX) was used to analyze the surface morphology and the chemical composition of the samples. Brunauer-Emmett-Teller (BET) method was carried out with nitrogen adsorption-desorption measurements at liquid nitrogen temperature (−196 °C) with an autosorb BET apparatus, Micromeritics ASAP 2020 in order to determine the textural property of the catalyst. Surface area and porosity analyzer were used to determine the surface area, pore size, and the pore volume.

2.4 Physico-chemical characterization of crude (jatroha) oil

Standard methods and procedures adopted by American Society for Testing and Material (ASTM and EN 14104) and that of official Methods of Analysis. (AOAC) 1997 were followed for acid value and all other parameters as reported in the work of Asuquo et al.

2.5 Transesterification process

All the transesterification processes were carried out in a 250 ml three necked round bottom flask equipped with a Liebig condenser and the thermometer as reported in previous work of Farooq et al. The raw materials involved in the reaction are jatropha oil, methanol and a catalyst (MgO/MCM-22). Initially, 12g of methanol and 0.72 g catalyst were mixed together in the flask connected to a reflux condenser to prevent the loss of methanol. The mixture was heated at different range of temperatures (40-80 °C) and stirred by magnetic stirrer at 500 rpm for several times (40-120 mins). Subsequently, 21.75 g of oil were added into the mixture and continued the reaction until the reaction is completed at definite period of time. Upon the reaction completion, phase separation was carried out in a separating funnel at room temperature. The influence of metal content (5-30 wt%) on the support (MCM-22) on the biodiesel yield was investigated by varying the catalyst amount (1-5 wt%), methanol to oil molar ratio (1:6-1:18), reaction time (40-120 mins), and temperature (40-80 °C). The range of experiment conducted are; 1-5 wt% (1, 2, 3, 4 and 5 wt%) catalyst loading, 1:6 to 1:18 (16, 1/1, 1,1/12, 1,1/5, and 1:18) methanol to oil molar ratio, reaction temperature of 40-80 °C (40, 50, 60, and 70 °C) and reaction time of 40-120 mins (40, 60, 80, 100 and 120 mins). The yield of biodiesel was determined. Then, the FAME yield was calculated according to equation (1). The weight of the product biodiesel was determined by weighing the FAME product with weighing balance after several washing with warm distilled water and sodium sulphate.

\[
\text{FAME yield} = \frac{\text{Actual weight (g)}}{\text{Theoretical weight (g)}} \times 100\% \quad (1)
\]

2.6 Physicochemical properties of produced FAME

The fuel properties of the synthesized methyl ester such as density and specific gravity, viscosity, pour point, cloud point, cold filter flogging point, acid value and other important parameters were measured using well established methods obtained from ASTM 4052 and EN6.

3. Results and Discussion

3.1 Catalyst testing and screening

In this analysis, the activity of all synthesized catalysts were tested in transesterification reaction at 1:15 molar ratio, 3 wt% catalyst loadings, 2 h of reaction time and at a temperature of 70 °C in order to determine the best catalyst. As can be seen in Fig. 1, 20MgO/MCM-22 (20 wt% MgO loading) gave higher yield than the other catalysts.

From the FAME yield obtained (Fig. 1), it can be found that, incorporation of MgO on MCM-22 improves the performance of the catalyst. FAME yield increases with increasing MgO loading up to 20 wt%. The enhanced catalytic activity of the MgO on MCM-22 catalyst might be connected to the existence of the optimal strength of the active basic and acidic sites on the surface of the catalyst to form the highly reactive methoxide species that can be reacted with triglycerides during the transesterification process. Based on the result obtained, 20 wt% MgO
loading (20MgO/MCM-22) appeared to be the best catalyst, hence, further study were based on it. It can be observed after 100 mins of transesterification reaction, the yield of FAME decreased. This is because excess reaction time did not promote forward reaction but caused soap formation which led to decrease in FAME yield. Ramadhas et al. 5) also reported similar observation while producing biodiesel from rubber seed oil.

3.2 Catalyst Characterization

3.2.1 XRD

The result revealed from the XRD shows the representative peak zeolite (Fig. 2). The diffraction peaks of MgO observed in MgO/MCM-22 can be matched with JCPDS file: 45-0946. The diffraction pattern of MCM-22 support shows peaks around 2θ = 22.2-24.9°, 26.8°, 30° and 45° that are characteristics of zeolite indicating typical zeolite structure 6). The XRD diffraction shows that MCM-22 possesses a highly amorphous structure in nature as compared to when MgO in incorporated. The increase in crystal shape of the MCM-22 after the successful intercalation of MgO sample is due to the good dispersion of MgO into the MCM-22 support 6). The major mineral found is silica (SiO2) which is a framework of silicon-oxygen hexagonal in shape. Other elements such as Na, Ca, and Al were also observed. The obtained results disclosed that the incorporation of little amounts of magnesium oxide (5-15 wt%) did not cause any serious change in morphology or showing any peaks related to crystalline MgO and the structure was preserved.

However, after the incorporation of high amount of MgO, the result revealed the existence of strong and sharp diffraction peaks located at the 2θ value of 32.8, 38.3 and 62.5° which correspond to (1 1 1), (2 0 0) and (2 2 0) planes, respectively indicating the successful formation of MgO 8). The diffraction peaks of MgO can be matched with standard Joint Committee on Powder Diffraction Standards-JCPDS data [JCPDS file: 45-0946]. This confirmed that, the addition of MgO at lower weight percent did not affect the structure of MCM-22, nevertheless, high loading affected the structure of MCM-22 which is in agreement with the previous work 7) 8).

3.2.2 BET

As earlier reported from the XRD results (Fig. 2), reduction in amorphous structure was observed in MgO/MCM-22 due to the effect of good dispersion of metals into the MCM-22. Proper calcination at the catalyst synthesis stage also has an effect on the textural properties of the catalyst. It can be seen from Table 1 that there is an increase in the pore volume and pore size of MgO/MCM-22 compared to the parent support (MCM-22). The increase in the amount of adsorbed N2 shown in the isotherm of MgO/MCM-22 as compared to that of MCM-22 in Fig. 3.
confirmed the increase in the pore volume. The samples displayed characteristics of Type II isotherms at lower pressure, which is a representative of the formation of monolayer and followed by multilayer\textsuperscript{9}. But, at high relative pressures, the isotherms ultimately resembles Type IV isotherm with the sharp uptake of N\textsubscript{2} which is highlighting the likely presence of large inter particle voids formed by agglomeration of the meso-like particles containing slit-shaped pores, which are representatives of H3 hysteresis loop\textsuperscript{9,10}.

This observation can be attributed to the solids having mesoporous shapes\textsuperscript{11} and this was validated by Fig. 4 for MgO/MCM-22 showing average pore size of 10.7 nm which falls within the 2-50 nm range for mesoporous materials. The obtained pore size of MgO/MCM-22 can be considered suitable for this study for transesterification reaction since previous study confirmed that mesoporous materials has higher catalytic activity and selectivity than the other materials\textsuperscript{11}, therefore, the catalyst pore can have unconstrained diffusion through the oil. Also it can be observed that the surface area decreased after the incorporation of metal oxides due to the partial blockage of the pores by the active metals during catalyst synthesis stage\textsuperscript{12,13}. It can also be observed that, the surface area of the catalysts was a bit low (24 m\textsuperscript{2}/g) compared with one reported in literature. However, in comparison with other heterogeneous catalyst and other silica, the surface area can be considered as high.

### 3.2.3 FESEM

FESEM was used to analyze morphology and size distribution of the MgO into MCM-22 support. The images of MgO with high magnification shows that MgO is successfully inserted into MCM-22. Fig. 5 displayed the morphology of the MCM-22 support before and after MgO loadings. The structure of the MCM-22 support revealed that, the catalyst is in highly amorphous form (as supported...
by XRD\(^{14}\). However, after the successful MgO loadings there was observable change in the initial structure of amorphous to a more of crystalline or variation in the morphology of the MCM-22 support as shown in Fig. 5 (b). This showed that, addition of high amount of metal oxide into the parent MCM-22 support was seriously tamper with the structural and textural integrity of the support.

Usually, as reported from previous study, aluminosilicates undergoes dealumination of framework of aluminum during catalysis synthesis due to thermal and protonation effects, which leads to the increase in Si/Al ratio, pore volume, and pore sizes as will be seen in Table 1\(^{15}\). The incorporation of MgO were seen to be highly dispersed on the MCM-22 support (Fig. 5 (b)) clearly due to the synthesis protocol where the metal oxide (MgO) precursor were drop-wisely added to the support dispersion under continuous vigorous stirring.

### 3.2.4 EDX

The result of elemental composition study of MCM-22 and that of 20 wt% MgO loading on MCM-22, carried out using EDX, are presented in Table 1 and Fig. 6 revealed that silica and alumina are the major components presents in MCM-22 with some trace of sodium, which show MCM-22 is a typical zeolite. The result is in high conformity with that of XRD findings. When MgO is incorporated into the MCM-22, the EDX shows the presence of Mg and O elements. The Figures and the Table demonstrates the presence of MgO in the prepared catalyst. It is confirmed

Table 1 Textural properties of MCM-22 and 20MgO/MCM-22

| Samples       | Total surface area (m\(^2\)/g) | Average pore size (nm) | Total pore volume (cm\(^3\)/g) | Elemental composition (%) |
|---------------|--------------------------------|------------------------|-------------------------------|---------------------------|
| MCM-22       | 23.5                          | 26                     | 0.015                         | Si 41.10 Al 49.91 Na 4.22 Mg 3.40 |
| 20MgO/MCM-22 | 17.6                          | 107                    | 0.047                         | Si 29.21 Al 58.08 Na 2.07 Mg 8.94 |

Fig. 5 FESEM images of (a) MCM-22, and (b) 20MgO/MCM-22

Fig. 6 EDX spectra of MCM-22 and 20MgO/MCM-22
that the grown nanoparticles are composed of Mg and O with trace of other metals, such as Na. The EDX also revealed that successful intercalation of MgO into the parent support of MCM-22 which increases the Si/Al ratio from 9.7 to about 14 as shown in Table 1. This increment was ascribed to dealumination of MgO/MCM-22 due to the effect of MgO activation and thermal treatment during catalyst synthesis stage and calcinations, respectively. Similarly, Pal-Borbely 15) reported reduction in the amount of zeolitic water under thermal treatment which is usually accompanied by an increase in the Si/Al ratio.

### 3.3 Physico-chemical properties of jatropha oil

The important properties of jatropha oil was characterized and the result is presented in Table 2. Standard procedures of American Society for Testing and Material (ASTM) and EN 14104 were followed for measuring saponification value, free fatty acid, and acid value determination by titration (ASTM standard 14104) as reported by Asuquo et al. 3). Color was observed from its physical appearance. Density and specific gravity were determined by density meter DMA 4500M. Viscosity, and moisture content were determined by CAP 1000+ viscometer, and halogen moisture content analyzer HR83, respectively.

All the obtained Physiochemical properties of the jatropha oil were found to be within the ASTM standard and can be compared well with the previous work from jatropha oil as well as other oils such rubber seed oil, castor and shear butter oils 3).

### 3.4 Optimization of transesterification reaction parameters

#### 3.4.1 Effect of catalyst loading on FAME yield

It was reported by Ramdhas et al. 5) that, transesterification reaction is inactive without catalyst. Acid or alkali catalyst in transesterification reaction helps to allow methanol to access the oil during transesterification process. The amount of catalyst used in the process also affects the FAME yield which is in agreement with the previous work. The catalyst weight percent was varied in the range of 1 to 5 wt% (1.0, 2.0, 3.0, 4.0 and 5.0). The result is shown in Fig. 7.

A careful observation from Fig. 7 shows that, at lower catalyst weight percent, there was a low yield of methyl esters. But as the weight percent increased, the yield increased where the highest yield of 98% FAME was achieved at 3 wt%. Further increase of the catalyst wt% resulted to a slight decrease in yield. As reported by Monteiro et al. 16), addition of excess amount of catalyst, gave rise to the soap formation which increased the viscosity and led to the formation of gels. It was also reported that 16), from physical appearance, the higher catalyst concentration, the darker the biodiesel produced, hence the addition of a proper catalyst is essential to the physical appearance of biodiesel.

#### 3.4.2 Effect of molar ratio on FAME yield

It was reported from the previous study that, transesterification reaction requires three moles of alcohol for each mole of oil 5). However, in order to drive the reaction towards completion, the molar ratio should be higher than reported stoichiometric ratio. Gerpan and Canakci 17) reported the use of large excess quantities of methanol about 1:15-1:35 while producing biodiesel from vegetable oil is very essential. Molar ratio is defined as the ratio of number of moles of alcohol to number of moles of triglycerides in the oil. In this work, various molar ratios of oil to methanol were tried starting with 1:6 until 1:18 (1:6, 1:9, 1:12, 1:15, and 1:18) and the result obtained were presented in Fig. 8.

As can be seen, when methanol/oil molar ratio increased from 16 to 1:15, there was an observed increase in the FAME yield (Fig. 8) where the maximum yield of 98% at molar ratio of 1:15 was attained. Consequently, as the molar ratio increased to 1:18 the yield dropped slightly to 82%. The decrease in the yield at the higher molar ratio may be connected to the low concentration of catalyst in the reaction system which was caused by excess methanol

| Parameters            | Value  |
|-----------------------|--------|
| Acid value (mg-KOH/g) | 3.61   |
| FFA (%)               | 1.805  |
| Water content (%)     | 0.06   |
| Density (g/cm³)       | 0.9124 |
| Iodine value (mg-I2/g)| 6.35   |
| Specific gravity at 20°C | 0.9158 |
| Kinematic viscosity (mm²/s)| 8.70   |

Fig. 7 Effect of catalyst wt% on FAME yield (Reaction time = 100 min, molar ratio = 1:15, tempt=70 °C)
in the reaction system. Fan et al.18 and Ramdhas et al.5 also
observed a similar decrease in the yield at high molar ratio.
Hence, molar ratio of 1:15 was considered to be the best
molar ratio in this study.

3.4.3 Effect of reaction temperature on FAME yield

Temperature is regarded as one of the most
important features that affect biodiesel production because
it can accelerate the rate of reaction by allowing methanol
to mix with oil and the catalyst and also allow the product
(FAME) out of the reaction mixtures 20) 21) . Also, it was
reported that, transesterification process is an endothermic
in nature 22) , therefore, for high FAME yield it may
require a high temperature. In this paper, five different
temperatures range from 40 to 80 °C were selected for the
transesterification reaction. Increase in FAME yield was
observed as the temperature rose from 40 °C until 70 °C
(Fig 9). The increased in the FAME yield as the
temperature increases was supported by the fact that,
increase in temperature is believed to facilitate the mixing
of the oil and the catalyst, thereby creating a void which
serves as migratory space for the reactants into the pore
and products out of the pore 23).

However, as can be seen in Fig 9, at reaction
temperature higher than 80 °C, a decreased in FAME
yield was observed and this may be attributed to the loss
of methanol during the reaction because boiling point of
methanol is less than 65 °C. Also, it was reported that,
high reaction temperature increase the production cost of
biodiesel 23).

3.4.4 Effect of reaction time on FAME yield

Reaction time is an important factor to consider
for transesterification reactions as lengthy agitation/heating
can cause soap production and degrade the heterogeneous catalyst 20). As reported by Ramdhas et al.5 that, in order to achieve good interaction between
the reagents (methanol and catalyst) and the oil during
transesterification reaction the mixture must be stirred
well at constant rate. Reaction time range between 40 until
120 mins were tried in this work and the results are shown
in Fig. 10. A clear observation from the Figure shows that,
the yield of FAME increases with the increase in reaction
time. Results achieved from the present work confirmed
that, about 100 min of reaction time is adequate enough
for transesterification reaction. What is typically and
frequently reported in the literature is that, heterogeneous
catalysts always display reduced activity and require longer
reaction times in order to give higher yield with respect to
their homogeneous counterpart due to less efficient mass
transfer, but the high activity observed in this work shows
that the low mass transfer might be positively balanced by
confinement and hydrophobic effects.

A similar trend was reported by Ramadhas et al.5 and
Ginting et al.24, while working on rubber seed oil and
jatropha seed oil, respectively.

3.5 Catalyst reusability test

The reusability of the catalyst is very essential
feature to be consider for determining the catalyst
sustainability and for commercial use. Fig. 11 shows the
result of three reusability studies on the MgO/MCM-22
catalyst for biodiesel production from jatropha oil with a
slight decrease from 98 to 90% after the first three runs.
The higher reusability of the catalyst can be attributed to its preparation procedures, especially the addition of MgO and the calcination. Furthermore, the ability of the MCM-22 to provide adequate anchor for the Mg ion made MCM-22 a favorable heterogeneous catalyst support for biodiesel production and other industrial application. It was also reported that, proper calcination has the ability to transform a metal oxide from amorphous form into crystalline thereby minimizing its toxic removal affinities.

3.6 Physico-chemical properties of FAME

The fuel properties of jatropha oil methyl ester in relationship with that of other esters is presented in Table 3. The majority of the properties of the produced FAME are reasonably comparable to those of other esters. The present result shows that, transesterification process improved the fuel properties of the oil with respect to viscosity and acid value. Most of the obtained properties fall within the range of biodiesel standard. The observed properties of methyl esters from jatropha oil were found to be in reasonable conformity with the ASTM standard.

The viscosity obtained in this work is lower than that obtained by Ginting et al. while working on jatropha oil as shown in Table 3. The acid value was found to be 0.62 mg-KOH/g. As reported, the low acid value of biodiesel indicates that, the biodiesels are not capable of causing corrosion to engine or storage tank. The obtained pour point, cloud, and cold filter plugging point indicate that the produced biodiesel can be used in temperate regions and is not capable of causing any vapor lock in diesel engines. Previous study reported that, in the conventional transesterification acid value and water always produce negative effects since the presence of high acid value and water causes soap formation, consumes catalyst, and reduces catalyst effectiveness. However, the results obtained from this study showed that, the produced FAME is within the acceptable limit of ASTM and EN for all parameters obtained and is well in conformity with the previous work from jatropha oil and other oils.

4. Conclusion

The present study demonstrates the successful incorporation of MgO by impregnation method into mesoporous MCM-22 aluminosilicates as an efficient catalyst for biodiesel production form jatropha oil. It was obtained that, the properties of the MCM-22 silica are affected by the incorporation of metal oxide as can be observed from the XRD results. It was also confirmed that, the prepared catalyst of MgO/MCM-22 show an increase in catalytic activity during transesterification reaction of jatropha oil compared with the parent support due to the strength of the active sites on the surface of metal oxide (MgO). It was discovered that, among the metal loadings prepared, 20 wt% loading showed a tremendous catalytic activity and gave higher yield of 98% than the rest of the catalysts. The optimum conditions were found to be 3wt% catalyst loading, 70 °C reaction temperature, 1:15 oil to methanol molar ratio and 100 mins reaction time. It can be concluded that, MgO incorporated into MCM-22 can serve an excellent solid catalyst for biodiesel production from the low grade jatropha oil through transesterification reaction. Moreover, the prepared catalyst was found to have a high chemical and thermal stability as it can be reused up to three times.
during transesterification without showing any major decrease in its catalytic activity.

Acknowledgment

The authors are grateful to the Yayasan Universiti Teknologi PETRONAS for the research funding and Universiti Teknologi PETRONAS for Graduate Assistance.

References

1) Rioux, R. M.; Song, H.; Hoefelmeyer, J. D.; Yang, P.; Somorjai G. A.; J. Phys. Chem. B. 109, 2192 (2005)
2) Farooq, M.; Ramli, A.; Duvvuri, S., Journal of cleaner Pro. 59, 131-140 (2013)
3) Asuquo, J. E.; Anusiem, A. C. I.; Etim, E. E., International Journal of Modern Chemistry 1, 109-115 (2012)
4) Abdulkadir, B. A.; Uemura, Y.; Ramli, A.; Othman, N.; Kusakabe, K.; Kai, T., J. Jpn Inst. Energy 94, 763-768 (2015)
5) Ramadhas, A. S.; Jayaraj, S.; Muraleedharan, C., Fuel 84, 335-340 (2005)
6) Yu, Y.; Xiong, G.; Li, C.; Xiao, F., Microp Mesop Mater 46, 23-34 (2001)
7) Jiang, D.; Zhao, B.; Xie, Y.; Pan, G.; Ran, G.; Min, E., Applied Catalysis A 219, 69-78 (2001)
8) Koo, K. Y.; Roh, H.; Jung, U. H.; Seo, D. J.; Seo, Y. S.; Yoon W. L., Catalysis Today 146, 166-17 (2009)
9) Xue, M.; Wang, Y., Microp Mesop Mater 156, 29-35 (2012)
10) Herney-Ramirez, J.; Vicente, M. A.; Madeira, L. M., Applied Catalyst B 98, 10-26 (2010)
11) Moniruzzaman M.; Sundararajan, P. R., Pure Applied Chemistry 76, 1335-1363 (2004)
12) Sum, O. S. N.; Feng, J.; Hu, X.; Yue, P. L., Chemical Engineering Sciences 59, 5269-75 (2004)
13) Dong, X.; Feng, L.; Ning, Z.; Fukui, X.; Junwei, W.; Yisheng, T., Appl. Catal. B: Environ., 191, 8-17 (2016)
14) Liu, X.; Wang, Y.; Wang, X.; Zhang, Y.; Gonh, Y.; Xu, Q.; Xu, J.; Deng, F.; Dou, T., China Journal of Catalyst 33, 1889-900 (2012).
15) Pal-Borbely, G., Molecular Sieves, 5, 67-101 (2007)
16) Monteiro, M. R.; Ambrozin, A. R. P.; Liao, L. M.; Ferreira, A. G., Talanta, 77, 593-605 (2008)
17) Canakci, M.; Garpen, V., Trans Am Soc Agric Eng. 42, 1203-1210 (1999)
18) Fan, Z.; Fang, Z.; Yi-Tong, W., Applied Energy, 155, 637-647 (2015)
19) Boocock, D. G. K.; Konar, S. K.; Mao, C.; Lee, C.; Buligan, S., Journal of the American Oil Chemists’ Society, 79, 1167-1172 (1998)
20) Hincapié, G.; Mondragón, F.; López, D., Fuel, 90, 1618-1623 (2011)
21) Yu, X.; Wen, Z.; Li, H.; Tu, J.; Yan, J., Fuel, 90, 1868-1874 (2011)
22) Chandrajai, S.; Prathima, B. K., Asian journal of chemistry, 16, 1811-1819 (2004)
23) Medy, C. N.; Tehirioua, E.; Lynda, E.; Kouassi, B. Y.; Erwan, L. G.; François-Xavier, F., Renewable Energy, 106, 135-141 (2017)
24) Ginting, M. S. A.; Azizan, M. T.; Yusup, S., Fuel, 93, 82-85 (2011)
25) Feng, J.; Hu, X.; Yue, P. L., Water Res., 40, 641-6 (2006)
26) Liu, F.; Asakura, K.; He, H.; Liu, Y.; Shan, W.; Shi, X.; Zhang, C., Catal Today, 164, 520-527 (2011)
27) Young, T.; Eom, C. Y.; Song, T.; Cho, J. W.; Kim, Y. M., Journal of Bacteriology, 179, 8 (1980)
28) Radovanovic, M.; Venderbosch, R. H.; Prins, W.; van Swaaij, W. P. M., Biomass Bioenergy, 18, 209-222 (2000)
29) Anjana, S.; Ram, P., Renewable and Sustainable Energy Reviews, 4, 111-133 (2000)