Catalysis Communications 183 (2023) 106749

Contents lists available at ScienceDirect

Catalysis Communications

journal homepage: www.elsevier.com/locate/catcom

New route for preparing palmitic acid imidazole from free fatty acid using imidazole

Adeeb Hayyan [a,*], Wan Jefrey Basirun [b,*], Muhammad Zulhaziman Mat Salleh [c,*], Mohamad Hamdi Zainal-Abidin [d], Waleed Al Abdulmonem [e], Rana Abdullah Alghamdi [f], Laila Alhussain [g], Abdullah S.M. Aljohani [h], M.Y. Zulkifli [i], Boey Min-Wei [a], Jehad Saleh [j], Fathiah M. Zuki [a], Mohamed E.S. Mirghani [k]

[a] Department of Chemical Engineering, Faculty of Engineering, Universiti Malaya, Kuala Lumpur 50603, Malaysia
[b] Department of Chemistry, Faculty of Science, Universiti Malaya, Kuala Lumpur 50603, Malaysia
[c] Department of Chemical and Process Engineering, Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia
[d] Department of Chemistry, Faculty of Science, Universiti Teknologi Malaysia, 81310 Johor Bahru, Malaysia
[e] Department of Pathology, College of Medicine, Qassim University, Buraydah 51452, Saudi Arabia
[f] Department of Chemistry, College of Sciences & Arts, King Abdulaziz University, Rabigh, Saudi Arabia, Saudi Arabia
[g] Department of Biology, College of Science, Qassim University, Buraydah 51452, Saudi Arabia
[h] Department of Veterinary Medicine, College of Agriculture and Veterinary Medicine, Qassim University, Buraydah, Saudi Arabia
[i] Halal Research Group, Academy of Islamic Studies, Universiti Malaya, Kuala Lumpur 50603, Malaysia
[j] Chemical Engineering Department, King Saud University, P.O. Box 800, Riyadh 11421, Saudi Arabia
[k] International Institute for Halal Research and Training (INHART), International Islamic University Malaysia, Kuala Lumpur 50728, Malaysia

ARTICLE INFO

Keywords:
Imidazole
Palmitic acid imidazole
Esterification
jatropha oil
Crude palm oil
Free fatty acid

ABSTRACT

In this study, imidazole was used for the first time in esterification of free fatty acid (FFA) from acidic oil and for palmitic acid imidazole production. The FFA content in jatropha oil mixed with crude palm oil (CPO) was significantly reduced from 10.57% to 1.73% under the following optimum conditions (25% imidazole dosage, 30 mins of reaction time, reaction temperature at 60 °C and methanol to oil molar ratio of 20:1). This research opens up new possibilities for utilizing imidazole as a catalyst in various esterification processes, offering a promising and eco-friendly pathway for industrial applications.

1. Introduction

One of the most common fatty acids in nature is palmitic acid, a saturated long-chain fatty acid [1]. Fatty acids and glycerol serve as vital raw materials, especially in oleochemical industries, and can be produced through the hydrolysis of fats and oils [2]. Hydrolysis is a chemical process that breaks down complex organic molecules into simpler monomers [3]. Being the largest palm oil exporters globally, Malaysia possesses its unique technology and formulation to ensure the production of high-quality palm oil [4]. The chemical esterification process, whether conducted with or without a catalyst, entails the separation of free fatty acids (FFA) from crude palm oil (CPO) under high-temperature conditions and within an inert medium [5].

The majority of palmitic acid (16 carbons) synthesis occurs either endogenously from other fatty acids, carbohydrate or from dietary intake [6]. It is frequently present in a variety of plant and animal oils, including palm oil and animal fats [7]. Fadzel et al. [8] reported that the content of palmitic acid and oleic acid in the range of 49 to 68% and 24 to 34%, respectively has been observed in various samples. Furthermore, a stearin variant with 79% palmitic acid content and 60% tripalmitoylglycerol (PPP) content is available, predominantly utilized as a firm base for soft margarine production and as a constituent in infant fat formulas. Moreover, palmitic acid is used in a variety of processes, including the synthesis of cellular membranes, energy storage, cosmetic ingredient and as a food additive [9,10].

Palmitic acid imidazole (PI) is a derivative of palmitic acid that holds significant interest in both industrial applications and academic research. It has one aliphatic chain, pentadecane, that links directly to the imidazole moiety and one palmitic unit that is attached to the imidazole moiety through an ethanamide unit. Due to its long aliphatic

* Corresponding author.
E-mail addresses: adeeb.hayyan@yahoo.com (A. Hayyan), jeff@um.edu.my (W.J. Basirun), zulhaziman@ukm.edu.my (M.Z. Mat Salleh).

https://doi.org/10.1016/j.catcom.2023.106749
Received 16 October 2022; Received in revised form 7 August 2023; Accepted 14 August 2023
Available online 16 August 2023

chain, heteroatoms, and π - bonds, PI has been used extensively as an effective corrosion-inhibiting molecule [11]. Imidazole derivatives have recently been reported to be both non-toxic and favorable to the environment [12]. Additionally, the water and oil solubility of the imidazole derivatives makes it simple to apply to a variety of applications [13]. In biological systems, imidazole derivatives possess a distinctive structural feature in the form of the imidazole ring, which exhibits desirable electron-rich characteristics. As a result, these derivatives can readily engage with numerous enzymes and receptors through a range of weak interactions, enabling versatile binding capabilities [14]. Previously, various imidazole analogues or derivatives have been reported to exhibit organocatalytic activities, particularly in hydrolysis-based chemical reactions [15]. Furthermore, imidazole has been discovered to accelerate the homogeneous esterification of starch over a limited number of cycles [16]. From the literature, it is evident that transesterification or esterification often utilizes polyhydric alcohols along with fatty acids derived from plants or animals [17].

Herein, in this study, imidazole is explored for the first time as an esterification catalyst for non-edible oils, which could be useful in converting low-grade oils into fatty acid imidazole or palmitic acid imidazole. This synthesis process requires careful consideration of reaction conditions, such as reaction temperature, catalyst dose, molar ratio, and reaction time, to achieve high yields. Furthermore, the study of molecular interaction of the resulting PI is essential to confirm its structure and verify its successful as an esterification catalyst. The research findings may shed light on new avenues for using this compound in areas such as pharmaceuticals, materials science and biotechnology.

2. Materials and methods

2.1. Raw materials and chemicals

The *Jatropha curcas* was obtained from local mills at Kordofan, west of Sudan while the crude palm oil (CPO) was obtained from local mill Selangor Malaysia. Methanol was acquired from Merck Sdn Bhd, Malaysia with purity of ≥99%. Imidazole ($C_3H_4N_2$) was purchased from Merck Sdn Bhd, Malaysia with purity of 99%. The other chemicals, i.e., methanol, potassium hydroxide (KOH), isopropanol and phenolphthalein were also purchased from Merck Sdn Bhd, Malaysia.

2.2. Methodology

2.2.1. Esterification

Jatropha curcas oil and CPO was mix with the volume ratio of 1:3 and the initial FFA content was 10.57%. This Acidic oil mixture was preheated to reduce the viscosity to facilitate the reaction. Jacketed reactor with reflux condenser control by feedback controlled system was utilized for the treatment of FFA content where the preheated oil (at 70 °C for one hour) samples were esterified using methanol with the presence of imidazole as catalyst. This method was adopted from a previous study [18].

The operating parameters for the initial experiment were 1% imidazole dosage, methanol to oil molar ratio of 10:1, 60 mins of reaction time, temperature at 60 °C, and stirring speed of 350 rpm. The presence of imidazole fatty acid was analyzed using Fourier Transform Infrared (FTIR) spectrometer (Nexus 670 Fourier Transform Infrared spectrometer, Thermo Nicolet, USA) and the FTIR spectra were analyzed using "Omnic 5.2a" software.

The FFA content after the esterification reaction was determined by a standard method Ca 5a-4 according to American Oil Chemists' Society (AOCS). The amount of active catalyst used in the reaction was calculated using Eq. (1). Catalyst consumption (C·C) is expressed as the mass of catalyst consumed per mass of synthesized product [18].

$$C.C = \frac{Mcat}{Mp} \tag{1}$$

where C·C = Catalyst consumption, Mcat = Mass of catalyst used to give the synthesized mass of product biodiesel (mg), Mp = Mass of synthesized product biodiesel (g).

The yield and FFA reduction were evaluated using titration according to the standard method Ca 5a-4 by AOCS. The transesterification reaction for treated oil was set at 1% of KOH solubilize in methanol (10:1, methanol: oil) at temperature of 60 °C, for 30 mins and stirring speed of 350 rpm [19].

$$Conversion = \frac{FFA_{IN} - FFA_{FIN}}{FFA_{IN}} \times 100\% \tag{2}$$

where FFA_{IN} and FFA_{FIN} are the initial and final FFA value of oil before and after esterification (g) respectively.

2.2.2. Optimization and reaction kinetics study

For the optimization of esterification of FFA, several parameters were applied in this study. The parameters and range of values for the optimization study are listed in Table 1 below.

The reaction kinetics were calculated for the optimum operating parameters after the optimization study. The order of reaction and rate of reaction, k was obtained by plotting graphs of concentration against time for all three kinetic models. The FFA content and conversion were determined to select the optimum operating condition for each parameter.

2.2.3. σ-profile and σ-potential

The geometry optimisation of all species involved in this study was performed using the Turbomole programme. The optimisation was performed at the Hartree–Fock level and 6-31G* basis set. The generation of *.cosmo* file was then conducted through a single-point calculation by using DFT with Becke–Perdew and the Triple-ζ Zeta Valence Potential (TZVP) basis set. Finally, the *.cosmo* files were exported to the COSMOthermX programme with parameterisation BP_TZVP_C30_1301.ctd. The σ-profile and σ-potential were generated from the global option of the COSMOtherm output files.

3. Results and discussion

3.1. Esterification of acidic oil using imidazole

3.1.1. Effect of imidazole dosage

The activation of imidazole showed a remarkable reduction in FFA content from 5.54% to 1.75% with the increased dosage from 1% to 30%. Fig. 1 shows that the FFA content decreases and conversion increases as catalyst dosage increases. As the limit or demand of final FFA after esterification was <2%, the optimum dosage of imidazole in esterification reaction was found to be at 25% as it reduces the FFA from 10.57% to 1.82%. As the dosage of catalyst increased beyond 30%, there was no significant or large increase in conversion, indicating an increase in C·C. Table 2 presents the effect of imidazole dosages on yield of treated acidic oil and C·C. At optimum conditions, the yield, C.C and treated oil conversion using 25% imidazole were 24.83 g, 302 mg/g and 82.78%.

Table 1
Range of parameters for optimization.

Parameter	Range
Imidazole dosage	1–50 wt%
Molar ratio (Methanol:Oil)	5–35: 1
Reaction temperature	35–80 °C
Reaction time	10–120 min

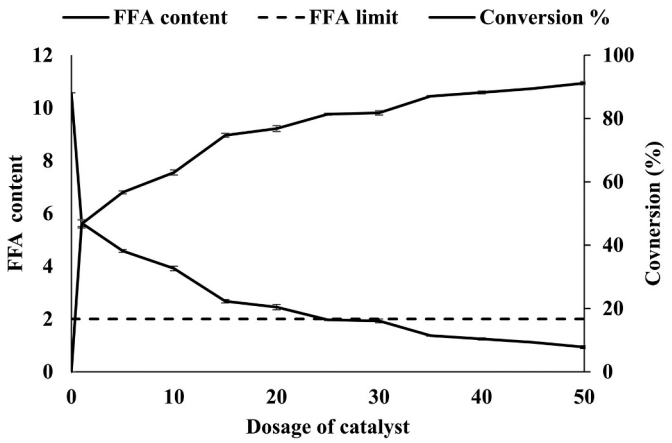

Fig. 1. Effect of imidazole dosage on FFA content reduction and the corresponding conversion of palmitic acid imidazole at 10:1 M ratio, 60 °C reaction temperature, 60 mins reaction time and 350 rpm.

Table 2
Catalyst consumption of imidazole in esterification reaction.

Dosage	Yield (gm)	C.C (mg/g)
1	14.27625	21.01392
5	17.14286	87.5
10	18.61873	161.128
15	22.59224	199.1834
20	23.33018	257.1776
25	24.83444	302
30	25.03311	359.5238
35	26.14002	401.683
40	26.33869	455.6034
45	26.87796	502.2703
50	27.19016	551.6701

3.1.2. Effect of molar ratio

The esterification reaction is a reversible reaction therefore excess methanol is needed to ensure the reaction shifts to the right. The reaction was performed by varying molar ratio of methanol to oil from 5:1 to 35:1, while maintaining the reaction at 60 °C and 25 wt% catalyst.

As shown in Fig. 2, the molar ratio of 20:1 (methanol:oil) resulted in 81 wt% of conversion and 1.92% of final FFA. However, this alcohol concentration did not lead to spontaneous phase separation. Despite the increase in methanol to oil molar ratio from 20:1 to 35:1, there was no apparent improvement in FFA reduction. This is because of the viscous nature of the fluid and accumulation of methanol. Therefore, in this study, the optimum molar ratio was set at 20:1.

3.1.3. Effect of reaction temperature

The reaction temperature is another crucial factor that affects the yield of treated oil. Fig. 3 shows the results of the experiments conducted at the optimal conditions of 25 wt% catalyst and 20:1 methanol to oil molar ratio, after 30 mins of reaction time at various temperatures between 30 and 80 °C. The high FFA content of acidic oil cannot be converted to 2% at temperatures below 50 °C. On the other hand, at 50 and 60 °C the FFA content was reduced to below 2%. This is due to the polarity of weak acid and methanol, that provides an increase in miscibility of acidic oil in methanol at 60 °C. The final FFA increased up to 2.3% for 70 °C and 2.5% for the reaction temperature of 80 °C, exceeding the FFA limit. This is probably due to the deactivation of the active acid as high temperature destructs the conformational structure of imidazole. Hence, the reaction occurring at 60 °C was found to be the most ideal.

3.1.4. Effect of reaction time

Fig. 4 illustrates the varying reaction time carried out at the optimal conditions of 25 wt% catalyst, 20:1 (methanol: oil) at temperature 60 °C. Esterification reaction times ranged from 10 to 120 mins while other parameters remained constant. The reaction achieved a steady state within 30 mins and reduced the FFA to <2%, resulting in 83.6% conversion. The FFA content gradually decreased away from the borderline of 2% after 40 mins and conversion decreased as the reaction time increased. After 120 mins of reaction time, 1.9% FFA content was achieved with 82% conversion. Therefore, the esterification reaction does not benefit from an increase in reaction time. The conversion of palmitic acid imidazole between 30 mins and 40 mins increased by only 1% and FFA content decreased by 0.11%, which is relatively insignificant. It was found that 30 mins are sufficient for esterifying FFA. Therefore, the optimal reaction time was determined as 30 mins.

3.2. Reaction kinetics

The kinetics analysis of the esterification using imidazole as catalyst were conducted by fitting the correspond data to zero order, 1st order, and 2nd order models. The linear fitting of the reaction kinetics model for the optimized operating parameters is as demonstrated in Fig. 5. Based on the parameters of the optimum reaction, the concentration of FFA was plotted against time. The order of reaction and reaction rate constant (k) were obtained from the kinetic model via the determination of coefficient of determination, R^2 from linear regression model. The R^2

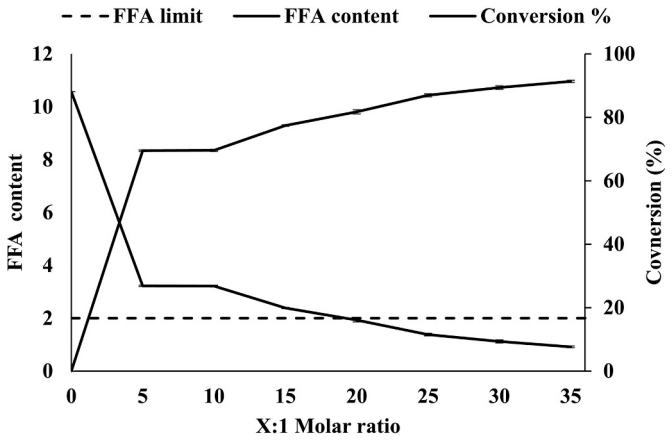

Fig. 2. Effect of molar ratio of methanol to oil on FFA content reduction and the corresponding conversion of palmitic acid imidazole at 25% imidazole dosage, 60 °C reaction temperature, 60 mins reaction time and 350 rpm.

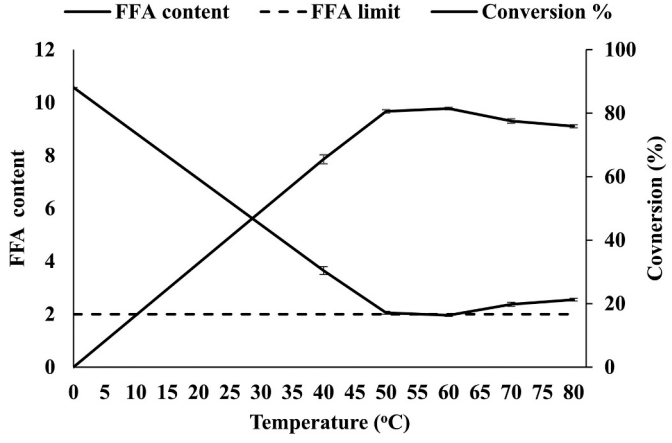

Fig. 3. Effect of reaction temperature on FFA content reduction and the corresponding conversion of palmitic acid at 25% imidazole dosage, at ratio 20:1 (methanol: oil), 30 mins reaction time and 350 rpm.

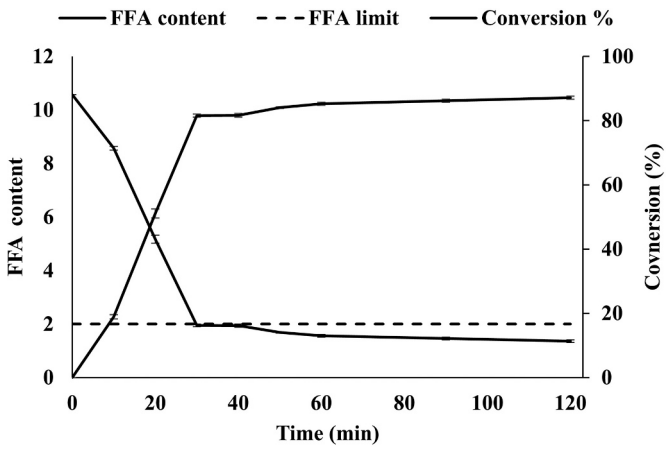

Fig. 4. Effect of reaction time on FFA content reduction and the corresponding conversion of palmitic acid imidazole at 25% imidazole dosage, at ratio 20:1 (methanol: oil), 60 °C reaction temperature and 350 rpm.

fits the second order reaction. In Fig. 5, the esterification facilitated by imidazole is illustrated as a second order reaction with a reaction rate constant of 0.0132 L mol^{-1} min^{-1}. The imidazole fatty acid was analyzed using FTIR technique. FTIR spectrum of imidazole shows peaks at 3125 cm^{-1} with a shoulder at 3123 cm^{-1} (Ring stretching). The imidazole heterocycle also presents two bands due to C = C and C = N stretch in the ring at 1574 & 1540 and 1479 & 1470 cm^{-1}. The spectrum for raw oil after treatment with imidazole shows a peak at 3420 cm^{-1} for the hydroxyl group –OH stretch and a shoulder peak of the fatty acid amide formed by the response at ~1644 for amide-I (C═O stretching) and 1240 cm^{-1} for amide-III. This is a strong indication of forming imidazole fatty acid after esterification reaction.

3.3. Validation

The effect of mixing device used in the esterification reaction was also investigated using different equipment. The experiment was conducted at different imidazole dosage, 20:1 (methanol: oil), at reaction temperature of 60 °C, and reaction time 30 mins. Shaker was set at 200 rpm whereas mechanical mixer was set at 350 rpm. The results of the experiments did not differ significantly when shakers or mechanical mixers were used (Fig. 6). It is due to the fact that imidazole is already soluble in methanol, which results in a homogeneous mixture. Since the reaction is homogeneous, the mixing device will not significantly affect the esterification reaction. Therefore, it is proven that the choice of equipment for mixing has no effects on the results obtained.

3.4. Molecular interactions

The molecular interactions between the species involved upon mixing can be qualitatively analyzed through the screening charge density (σ), or more specifically, σ-profile, σ-potential and σ-surfaces. In σ-profile and σ-potential, when the screening charge density exceeds ±0.0084 $eÅ^{-2}$, the molecule is considered sufficiently polar to induce hydrogen bonding. A higher absolute value of σ leads to a stronger compound as a hydrogen bond donor (HBD) or a hydrogen bond acceptor (HBA). On the other hand, the σ-potential indicates the affinity of a component in a mixture towards another. In the σ-potential plot, a higher negative value of μ (σ) indicates an increasing interaction between molecules, whereas a higher positive value signifies an increase in repulsive tendency. The σ-profile, σ-potential and σ-surface of imidazole, methanol and FFA (e.g: palmitic acid) are depicted in Fig. 7.

Palmitic acid clearly showed a high peak area in the non-polar region which is contributed by the long alkyl chain. The peak at 0.012 $eÅ^{-2}$ of palmitic acid belongs to the oxygen atom with two lone pair electrons of the carbonyl group (C=O). Upon mixing palmitic acid with imidazole and methanol, the intermolecular attractions occur between the hydrogen bond donating (blue σ-surface) and hydrogen bond accepting (blue σ-surface) regions of each species. In σ-potential plot, the lowest μ value is observed in imidazole (in HBD region) and palmitic acid (in HBA region) curves. This indicates a strong attraction between them, which is expected to induce atom transfer (esterification reaction), as experimentally shown by the reduced FFA content.

On the other hand, based on the σ-potential and σ-surfaces, methanol is expected to act only as the solvation media. During solvation, imidazole and palmitic acid are surrounded by concentric shells of methanol molecules, forming solvation complexes. Based on this analysis, the proposed reaction mechanism is depicted in Fig. 8. There is an affinity for methanol to interact with imidazole via partial charge attraction, but due low attraction energy, no transfer of electron or atoms can be expected. The products of this reaction are palmitic acid imidazole and water.

4. Conclusions

This work presents a novel method for esterifying FFA using a pharmaceutical-based catalyst, imidazole and to prepare palmitic acid

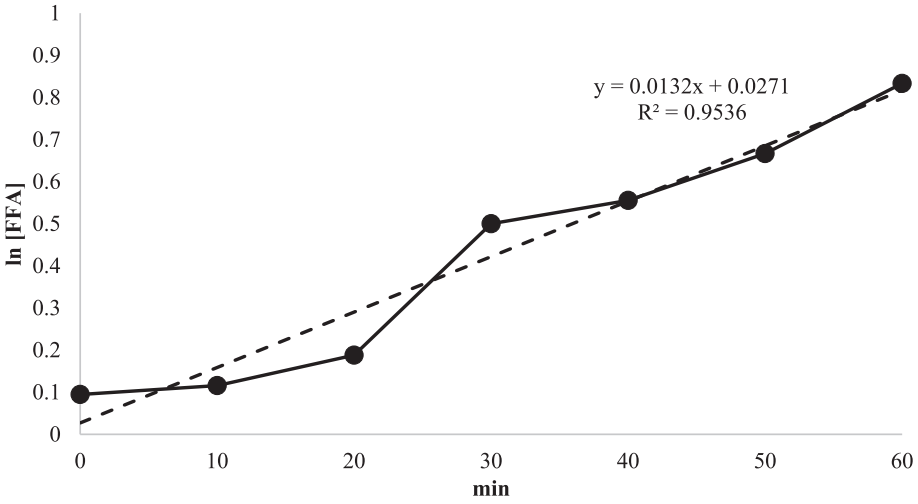

Fig. 5. Linear fitting of FFA concentration against time at 25% imidazole dosage, at ratio 20:1 (methanol: oil), 60 °C reaction temperature, 60 mins reaction time and 350 rpm.

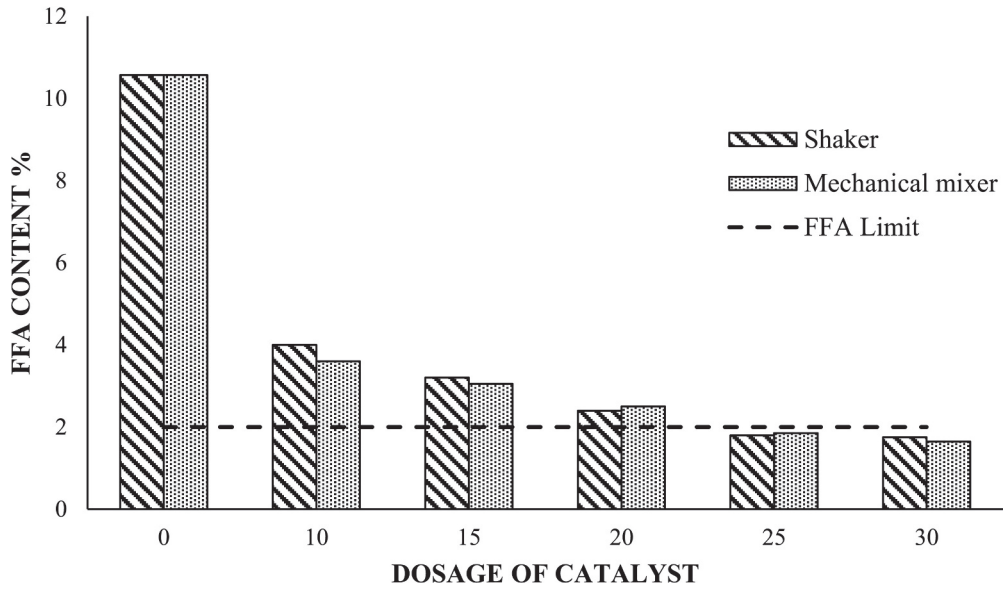

Fig. 6. Effect of equipment used on the reduction of FFA content and the corresponding conversion of palmitic acid imidazole at different imidazole dosage, at ratio 20:1 (methanol: oil), 30 mins reaction time, 60 °C reaction temperature and 350 rpm.

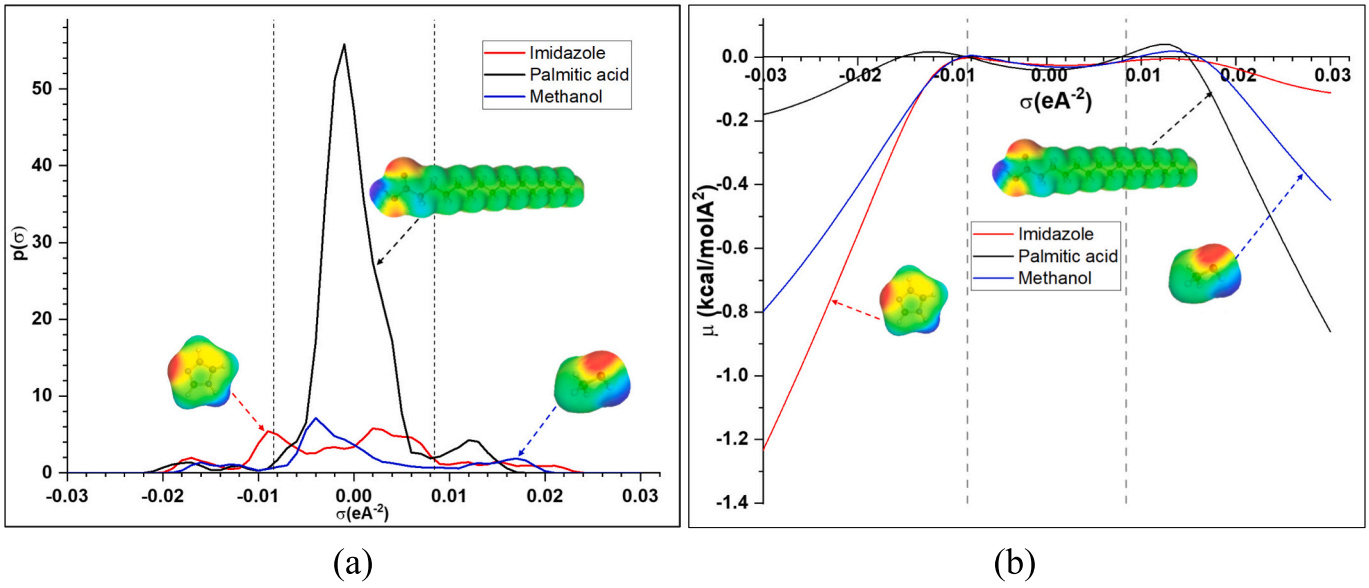

Fig. 7. Screening charge densities of imidazole, methanol and FFA (palmitic acid) in view of (a) σ-profile and (b) σ-potential.

imidazole. The optimization and kinetics studies were carried out and the results were obtained. Results showed that the optimized operating parameters significantly reduced the acidity of acidic oil from initial FFA (10.57%). The optimum operating conditions for the esterification of FFA with imidazole catalyst was found to be 25% imidazole dosage, 30 mins of reaction time, at temperature 60 °C and methanol to oil molar ratio of 20:1. The esterification reaction fits the 2nd order model and reaction rate constant was 0.0132 L mol^{-1} min^{-1}. Molecular interaction and proposed mechanism were presented in this study. Results proved imidazole to be a simple, cheap, and active alternative green catalyst for esterification reaction and for production of palmitic acid imidazole. This may enable the use of other pharmaceutical-based materials in other chemical reactions as well.

CRediT authorship contribution statement

Adeeb Hayyan: Conceptualization, Investigation, Data curation, Supervision, Writing – original draft. **Wan Jefrey Basirun:** Writing – original draft. **Muhammad Zulhaziman Mat Salleh:** Writing – original draft, Software, Data curation, Funding acquisition. **Mohamad Hamdi Zainal-Abidin:** Writing – original draft. **Waleed Al Abdulmonem:** Resources. **Rana Abdullah Alghamdi:** Resources. **Laila Alhussain:** Resources. **Abdullah S.M. Aljohani:** Resources. **M.Y. Zulkifli:** Funding acquisition. **Boey Min-Wei:** Writing – original draft. **Jehad Saleh:** Resources. **Fathiah M. Zuki:** Resources. **Mohamed E.S. Mirghani:** Formal analysis, Data curation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

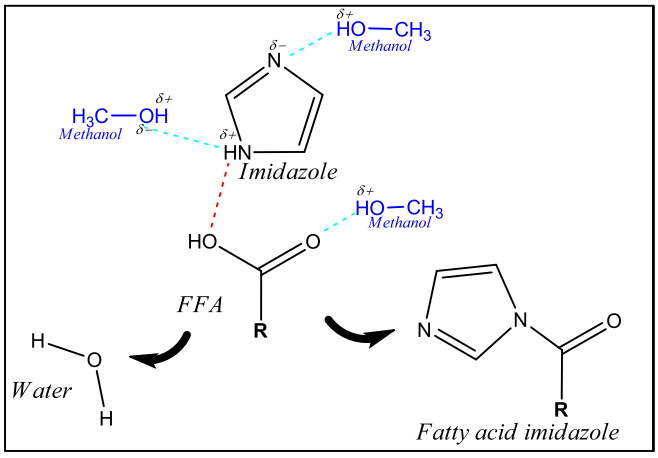

Fig. 8. Esterification reaction of FFA using imidazole in methanol.

Data availability

Data will be made available on request.

Acknowledgements

The authors express their acknowledgements to Universiti Kebangsaan Malaysia for all the supports under grant No. GGPM-2021-010.

References

[1] M.F. Khan, M.A. Quraishi, M. Maslehuddin, Z.M. Gasem, Corrosion inhibition of reinforcing steel using environment-friendly palmitic-acid-imidazole, SSRN Electron. J. (2022), https://doi.org/10.2139/ssrn.4009590.
[2] M. Bahadi, M.F. Yusoff, J. Salimon, D. Derawi, Optimization of response surface methodology by d-optimal design for alkaline hydrolysis of crude palm kernel oil, Sains Malaysiana. 49 (2020) 29–41, https://doi.org/10.17576/jsm-2020-4901-04.
[3] Siti Kartom Kamarudina, A. Al Muniandy, N.S. Shamsul, H.T. Kofli, Enhanced biogas production from agro wastes by co-digestion with crude glycerol, J. Kejuruter. SI1 (2018) 47–57, https://doi.org/10.17576/jkukm-2018-si1(1)-07.
[4] Zuhaira Mohd Zulkifli, F.H. Hashim, T. Raj, A.B. Huddin, A rapid and non-destructive technique in determining the ripeness of oil palm fresh fruit bunch (FFB), J. Kejuruter. 30 (2018) 93–101, https://doi.org/10.17576/jkukm-2018-30(1)-12.
[5] M.A. Bahadi, A.W. Japir, N. Salih, J. Salimon, Free fatty acids separation from Malaysian high free fatty acid crude palm oil using molecular distillation, Malaysian J. Anal. Sci. 20 (2016) 1042–1051, https://doi.org/10.17576/mjas-2016-2005-08.
[6] A.M.M. Ortega, M.R.S. Campos, Chapter 3 - Macronutrients and micronutrients in cancer prevention and treatment, in: Oncol. Funct. Nutr., Academic Press, 2021, pp. 99–124, https://doi.org/10.1016/B978-0-12-819828-5.00003-6.
[7] R. Chowdhury, M. Steur, P.S. Patel, O.H. Franco, Chapter 10 - individual fatty acids in cardiometabolic disease, in: Handb. Lipids Hum. Funct., AOCS Press, 2016, pp. 207–318, https://doi.org/10.1016/B978-1-63067-036-8.00010-X.
[8] F.M. Fadzel, J. Salimon, D. Derawi, Low-energy separation technique on purification of unsaturated fatty acids of palm stearin using methanol crystallization method, Sains Malaysiana. 50 (2021) 151–160.
[9] X. Jiang, X. Zou, Z. Chao, X. Xu, Preparation of human Milk fat substitutes: a review, Life. 12 (2022), https://doi.org/10.3390/life12020187.
[10] Q. He, Y. Chen, Z. Wang, H. He, P. Yu, Cellular uptake, metabolism and sensing of long-chain fatty acids, Front. Biosci. - Landmark. 28 (2023), https://doi.org/10.31083/j.fbl2801010.
[11] A. Dutta, S.K. Saha, U. Adhikari, P. Banerjee, D. Sukul, Effect of substitution on corrosion inhibition properties of 2-(substituted phenyl) benzimidazole derivatives on mild steel in 1 M HCl solution: a combined experimental and theoretical approach, Corros. Sci. 123 (2017) 256–266, https://doi.org/10.1016/j.corsci.2017.04.017.
[12] D.B. Tripathy, M. Murmu, P. Banerjee, M.A. Quraishi, Palmitic acid based environmentally benign corrosion inhibiting formulation useful during acid cleansing process in MSF desalination plants, Desalination. 472 (2019), https://doi.org/10.1016/j.desal.2019.114128.
[13] I.B. Obot, U.M. Edouk, Benzimidazole: small planar molecule with diverse anti-corrosion potentials, J. Mol. Liq. 246 (2017) 66–90, https://doi.org/10.1016/j.molliq.2017.09.041.
[14] H. Qin, Z. Song, Q. Zeng, H. Cheng, L. Chen, Z. Qi, Bifunctional imidazole-PTSA deep eutectic solvent for synthesizing long-chain ester IBIBE in reactive extraction, AICHE J. 65 (2019) 675–683, https://doi.org/10.1002/aic.16472.
[15] P. Nieri, S. Carpi, S. Fogli, B. Polini, M.C. Breschi, A. Podestà, Cholinesterase-like organocatalysis by imidazole and imidazole-bearing molecules, Sci. Rep. 8 (2017) 1–8, https://doi.org/10.1038/srep45760.
[16] S. Blohm, T. Heinze, Mechanistic considerations of efficient esterification of starch with propionic anhydride/Lauric acid in the green solvent imidazole, Macromol. Chem. Phys. 221 (2020) 1–10, https://doi.org/10.1002/macp.202000264.
[17] M. Bahadi, J. Salimon, D. Derawi, Synthesis of ISO grade 46 and 68 biolubricant from palm kernel fatty acids, Sains Malaysiana. 51 (2022) 2507–2529, https://doi.org/10.17576/jsm-2022-5108-13.
[18] A. Hayyan, M.A. Hashim, M. Hayyan, Application of a novel catalyst in the esterification of mixed industrial palm oil for biodiesel production, Bioenergy Res. 8 (2015) 459–463, https://doi.org/10.1007/s12155-014-9502-0.
[19] A. Hayyan, M. Ali Hashim, F.S. Mjalli, M. Hayyan, I.M. AlNashef, A novel phosphonium-based deep eutectic catalyst for biodiesel production from industrial low grade crude palm oil, Chem. Eng. Sci. 92 (2013) 81–88, https://doi.org/10.1016/j.ces.2012.12.024.