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Comparison of neural network and response surface methodology techniques on optimization of biodiesel production from mixed waste cooking oil using heterogeneous biocatalyst

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ABSTRACT

The present research aims to optimize the process parameters of biodiesel production from mixed waste cooking oil, consisting of sunflower and palm oil. The biocatalyst was synthesized from Chinese broccoli, Cauliflower and Napa cabbage vegetable wastes and potassium hydroxide-impregnated biocatalyst was synthesized through the incipient wet impregnation method. Artificial neural networks (Scaled Conjugate Gradient, Bayesian Regularization and Levenberg Marquardt Neural Network) and RSM-based central composite design are used to optimize the parameters in biodiesel production. The obtained predicted results from ANN and RSM models are compared with experimental results. The experimental results showed that biocatalyst impregnated with KOH has produced a maximum yield of 94.7 % at a reaction temperature of 60 °C, time of 120 min, catalyst loading of 7 wt%, 1:9 M ratio and 900 rpm stirrer speed. In addition, among the three algorithms of ANN, BRNN neural network had better prediction results in biodiesel production. At the same time, LMNN, and SCGNN are nearly similarly efficient on the other hand the BRNN model has lower error values. The ANN and CCD models have demonstrated lower RMSE and MAPE such as 0.001 to 0.018, and 3.22–4.37 % respectively, and high R² values in the ranges of 0.97 to 0.99, indicating the excellent reliability of these two approaches for process optimization. Hence, the above study recommended a biocatalyst impregnated with KOH as a catalyst to achieve maximum biodiesel yield. Further, ANN and RSM techniques are suggested for biodiesel process optimization and yield prediction.

1. Introduction

The world's energy demand is escalating every day due to industrialization, population growth, and technological development. Roughly, 80 % of fossil fuels and 20 % of renewable sources could meet the global energy demand. Oil accounts for 35 % of total energy output, followed by natural gas (24 %), coal (29 %), hydropower (6 %), and nuclear energy (5 %), and the remaining comes from solar and supplementary sources [1]. Global warming, climate change and environmental

pollution are caused by the constant increase in emissions that come from the combustion of fossil fuels [2]. US energy data show that the world's energy consumption is rising at a rate of 1.6 % annually. But, fuel prices and demand for petroleum fuel, scientists and researchers are looking for new renewable energy sources for diesel engines [3]. Among the renewable resources, biodiesel has a predominant alternative source due to lesser investment cost, maintenance and production cost, also it is environmentally friendly, biodegradable, and non-toxic compared to other resources. Most of the biodiesel is produced from edible and non-edible oil such as sunflower oil, palm oil, rapeseed oil and ground nut

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Nomenclature

ASTM	American Society for Testing and Materials
BRNN	Bayesian Regularization Neural Network
BBD	Box- Behnken Design
CaCO ₃	Calcium carbonate
CCD	Central Composite Design
DSC	Differential Scanning Calorimetry
FTIR	Fourier Transform Infrared Spectroscopy
GC-MS	Gas Chromatography and Mass Spectrometry
KOH	Potassium hydroxide
LMNN	Levenberg Marquardt Neural Network
FAME	Fatty Acid Methyl Ester
MWCO	Mixed Waste Cooking Oil
NaOH	Sodium hydroxide
RSM	Response Surface Methodology
SCGNN	Scaled Conjugate Gradient Network

oil, Jatropha oil and Karanja [4]. The production of biodiesel from vegetable oil destroys soil, and forests and uses more arable land. Additionally, the price of vegetable oil will rise, endangering the long-term viability of biodiesel. Further, it creates social conflict between food vs fuel. Hence, biodiesel production from edible and non-edible oil is not the right choice. After cooking, vast quantities of cooking oils have been dumped in drainage or land and can be used as feedstock in biodiesel production [5].

The drawbacks of homogeneous catalysts in biodiesel production include purification, and glycerol separation, whereas heterogeneous catalysts are free from the above-stated problem. Moreover, a higher reaction rate and conversion percentage when compared to homogeneous catalysts [6]. Xie and Huang et al. [7] synthesized a composite as an efficient and recyclable biocatalyst for biodiesel production from soybean oil. The Fe₃O₄-POLY (GMA-co-MAA) composites were characterized by FTIR, XRD, SEM, TEM, XPS and nitrogen absorption-desorption techniques. The maximum biodiesel yield of 92 % was obtained for magnetic biocatalyst at 40 °C temperature with three-step methanol injection. In another study, Zulfiqar et al. [8] developed a novel nanocatalyst (Lipase-PDA-TiO₂-NPs) for enzymatic transesterification of Jatropha curcas seed oil in biodiesel production. The TiO₂-NPs were prepared using a hydrothermal method and modified by the polydopamine polymer. The optimal biodiesel yield of 92 % was achieved at 57 °C with a 10 wt% of biocatalyst concentration and a 1: 6 M ratio. Takeno et al. [9] extracted a biocatalyst from silver croaker's stone. It was found that the stone is composed of calcium carbonate and the catalyst was obtained by calcination of calcium oxide. Also, the parameters were optimized by the Box-Behnken method. The study unveiled that the optimum yield of 97 % was achieved at 67 °C, 5.3 wt%, 175 min and 11:1 M ratio.

Numerous research works have been carried out on the optimization of biodiesel production from various types of feedstocks. Among those studies, Taguchi and RSM are the most used methods in biodiesel optimization. Although each method has a unique approach and set of challenges, they are maximizing the biodiesel production criteria [10]. It has been used in numerous fields of scientific research, including biofuel production, CO₂ capture, the petroleum refining sector and food engineering [11]. Rokni et al. [12] extracted biodiesel from Camelina oil through a microwave-assisted transesterification process, and the Box-Behnken method was employed to optimize the parameters. The studies found the optimum biodiesel yield of 95.31 % was obtained at a catalyst loading of 1.26 wt%, the molar ratio of 6.91, and the time of 5.85 min. In another research, Yusuff et al. [13] developed a barium-modified zeolite catalyst by using a co-precipitation technique followed by thermal treatment for biodiesel production from waste frying

oil. The biodiesel process conditions were optimized using a central composite design. The study revealed a supreme yield of 92 % was achieved at 3 wt% catalyst concentration, 1:12 M ratio, and a temperature of 65 °C. Anvita Sharma et al. [14] synthesized the biodiesel production from waste cotton seed oil using KOH and CaO as a catalyst. The study reported that experimental results were in good agreement with model prediction. The biodiesel yield of 96 % and 90 % were obtained for KOH and CaO respectively [14]. Balaji and Niju [15] synthesized a novel, inexpensive, environmentally friendly and highly effective heterogeneous catalyst from Musa acuminate Colla (Red banana) for biodiesel production. The study found that biocatalyst has maximised the biodiesel conversion yield of 98 % at 106 min of duration, 2.68 wt%, 1:12 M ratio and temperature of 65 °C, this is due to its massive multi-minerals and large surface area. Gunindra Pathak et al. [16] synthesized biodiesel from soybean oil using a heterogeneous catalyst made from banana peel ash, a waste biomass product. The catalyst was analyzed to determine its chemical composition, structure, and morphology through XRD, XPS and TEM. The optimum reaction condition led to a high biodiesel production of 98 %. This is due to metals present in heterogeneous catalysts serving as the primary sites for the transesterification reaction that make biodiesel. Further, it is a green catalyst for low-cost biodiesel production and has the advantage of being a waste material, biogenic, highly efficient and ecologically benign. Victoria et al. [17] optimized the biodiesel production from esterified palm kernel oil using a bio-based heterogeneous catalyst. A central composite design was employed to examine the individual and interaction effects of process variables. The study found that the CBPA catalyst produced the maximum yield of 99.5 % at a temperature of 65 °C, a molar ratio of 1:8, catalyst loading of 4 wt% and a time of 65 min. Methanol/oil had the greatest impact on biodiesel yield. Also, the RSM model offered a good prediction with experimental results with an R² value of 0.967.

Artificial neural networks (ANN) and response surface methodology (RSM) have proven to be dependable among other optimization strategies because of their precision and minimum error proportion [18]. Vinoth et al. [19] employed the ANN technique to optimize the parameters in biodiesel production from Aegle Marmelose Correa seed. The results revealed that the LMNN has exactly forecasted the biodiesel production with a greater R² value. Neeta Kumari et al. [20] used three different algorithms like the BRNN, the SCGNN and the LMNN network for the optimization of pretreatment process parameters. It was found that among the three algorithms, the LMNN model has more accurate prediction results with higher R² values and lower MSE. Vinoth Arul Raj et al. [21] optimized the biodiesel process parameters by using ANN and RSM techniques. the backpropagation network was analyzed using the Levenberg-Marquardt algorithm. The study reported that the R² value of ANN and RSM was found to be 0.97 and 0.87 respectively. Zheng et al. [22] used ANN and CMA-ES methods to optimize the engine emissions characteristics fuelled with biodiesel-diesel blends. The back-propagation Multilayer perception network model was used due to high precision and more accuracy than any other network model. Also, the study reported that the R² value, MSE and RMSE was found to be 0.98, 6.48 % and 3.2 ppm, respectively.

According to earlier studies, utilizing vegetable oil and non-edible oil in biodiesel production causes food vs fuel conflict, deforestation and soil erosion. Around 70 % to 80 % of biodiesel production costs are based on raw material costs. Among the various feedstock, MWCO is easily available and low cost and does not cause a problem between food and fuel, but it should be pretreated before being disposed into the environment to avoid soil or environmental pollution. The cost required for a pretreatment process is high resulting in direct disposal into land causing environmental pollution. Therefore, the mixed waste cooking could be converted into biodiesel in this research work. Several problems arise when using a homogeneous catalyst in biodiesel production including the inability to recover the catalyst, the need for expensive purification, and catalyst removal steps, equipment corrosion and



Fig. 1. Photographic view of the a) biodiesel setup, b) pretreated oil and c) biodiesel as shown in.

wastewater generation. As a result, heterogeneous catalysts were used in biodiesel production because it is environmentally friendly, economically viable, and increases the effectiveness of the transesterification process. Biomass-based heterogeneous catalyst has received more attention recently because of their availability and catalyst activity for the production of biodiesel. Due to its widespread availability on the local market and high mineral content, especially calcium and potassium, the waste of Chinese broccoli, Cauliflower and Napa cabbage has been used in this study to synthesis an eco-friendly heterogeneous biocatalyst for biodiesel production. Only a few papers on the statistical optimization of the transesterification process using a heterogeneous catalyst made from lignocellulose biomass. Additionally, there are not many publications about the production of heterogeneous catalyst-assisted biodiesel from mixed waste cooking oil. To increase the biodiesel yield, the central composite design of RSM and ANN was used in this study to optimize the transesterification reaction parameters such as reaction time, catalyst concentration, methanol to oil ratio, stirrer speed and reaction duration. Hence, an effort has been made to explore the influence of biocatalysts on biodiesel production from MWCO. Further, the biodiesel process parameters were optimized by using ANN and RSM approaches.

2. Materials and method

2.1. Biocatalyst synthesis and characterization

A large quantity of biomass waste is generated every year, and this is simply disposed to open environment or burned in the agricultural field. Hence, vegetable waste can be converted into biocatalysts for biodiesel production in this present work. Vegetable wastes such as Naga cabbage, Cauliflower and Chinese were procured from North Bangkok, Thailand. Further, absolute ethanol and Potassium hydroxide were bought from Sigma Aldrich, Thailand. In this study, the potassium hydroxide-impregnated biocatalyst was synthesized by the incipient wet impregnation method [19,23]. The catalyst was synthesized with the help of a tubular furnace (CHAVACHOTE, Thailand), an inner diameter of 5 cm and a maximum operating temperature of 1200 °C. The phase structure before and after the reaction of the samples was studied by X-ray diffraction (XRD) (Smartlab XE, Rigaku with D/teX Ultra 250), scan range $2\theta = 5-90^\circ$ with speed $10.00^\circ/\text{min}$, increasing step width 0.01° ,

Table 1

Experimental matrix for transesterification reaction at various conditions.

Molar ratio % v/v	Catalyst wt. %	Temperature (°C)	Time (minutes)	Speed (rpm)	FAME yield (%)
9	7	60	120	600	90
3	5	65	150	300	77
9	7	60	120	900	94
6	9	50	60	600	84
9	7	55	90	600	92
3	5	45	30	300	78
9	9	60	60	600	91
12	3	65	120	300	82
3	1	55	30	1500	73
12	7	45	120	1200	76
9	7	55	60	600	92
15	3	65	30	1200	74
12	7	45	90	1500	81
6	5	60	90	900	91
12	3	65	150	300	81
6	7	55	60	900	92
3	1	45	90	1200	73
6	5	50	90	900	86
12	1	45	150	1500	73
6	9	50	120	600	89
9	7	60	30	600	92
6	3	50	90	900	83
15	5	50	60	600	81
3	9	65	150	1200	76
3	1	45	150	300	72
15	9	60	60	900	87
15	5	50	90	600	82
3	3	45	120	1200	73
12	5	50	30	300	75
15	7	50	120	900	84
12	1	55	150	1500	73
3	9	65	120	300	81

using CuK α radiation ($\lambda = 1.542 \text{ \AA}$) and X-ray generator of 40 kV, 100 mA. Further, Scanning Electron Microscopy (Carl Zeiss Microscopy Ltd, UK) is used to examine the catalyst's morphology and lattice structure. It was operated at 200 kV of voltage.

2.2. Biodiesel synthesis from mixed waste cooking oil using biocatalyst

Mixed cooking oil consisting of sunflower and palm oil was acquired from restaurants in Bang Sue, Thailand. Waste cooking oil was purified by using a centrifuge running at 5000 rpm for 2hr to remove water and solid particles. Further, the MWCO was heated at 110 °C for 30 min before being poured into the reactor to remove the moisture content. The acid value of MWCO was found to be 3.14 mg KOH/g. It should be <1 mg KOH/g for the transesterification process. Hence, an esterification reaction was performed for MWCO under the following conditions, 1 % phosphoric acid, 55 °C, 90 min of reaction time, 1:3 M ratio and a stirrer speed of 900 rpm. As a result, the acid value of WCO was lowered to 1 mg KOH/g. A photographic view of the biodiesel experimental setup, waste cooking oil and biodiesel as shown in Fig. 1. A 200 ml round bottom flask with a heating mantle, thermocouple, condenser and magnetic stirrer was used in the transesterification process. To achieve homogeneous mixing, 100 g of mixed waste cooking oil, methanol, and potassium hydroxide impregnated with biocatalyst were mixed in a conical flask before being added to the reactor, and the mixture was placed into the round bottom flask and stirred for a given reaction time specified in the experimental matrix. The product was poured into the gravity-separating funnel to settle for 24 hr. The glycerol settled at the bottom side of the funnel, whereas biodiesel was placed on the top side of the separating funnel. The raw biodiesel has been purified by using distilled water and dried at 110 °C to remove water, unreacted catalyst and methanol. The presence of methanol or unreacted catalyst could reduce the flash point and calorific value of the biodiesel.

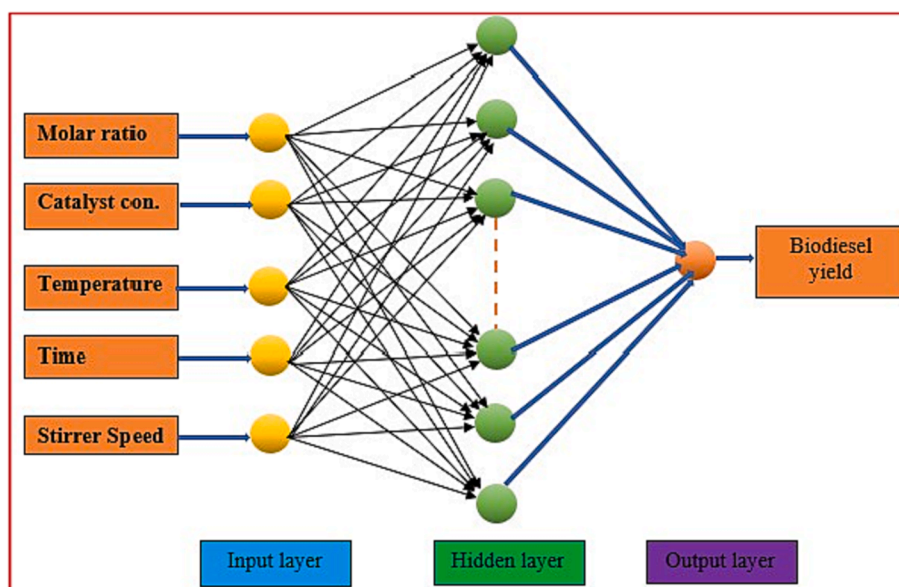


Fig. 2. ANN optimal architecture for the biodiesel production model.

2.3. Response surface methodology

Response surface methodology is a cost-effective, modelling and optimization tool for research and industry. It has been used in many fields of scientific research including biofuel production, food engineering, CO₂ capture and the petroleum refining sector [24]. In this present work, experimental design, modelling and process optimization work will be carried out by using an RSM-based central composite design (Design-Expert software version 10.0.3). A fixed number of tests are required for the CCD design, which is a five-level factorial design. In this work, time (X_1 , 30–150 min), temperature (X_2 , 45–65 °C), catalyst concentration (X_3 , 1–9 wt%), the molar ratio (X_4 , 1:3 to 1:15) and stirrer speed (X_5 , 300 to 1500 rpm) were designated as input parameters and biodiesel yield (Y , %) was output response [25]. The central composite design matrix included 31 experimental runs is depicted in Table 1.

2.4. Selection of input and output variables and modelling of ANN network

The artificial neural network works like a human brain neurological process, which eradicates the limitation of RSM and is a superior model since can accurately forecast a non-linear process throughout the optimization process [26]. Even from complex and vague data, ANN may perfectly identify the pattern of the human brain or conventional computers or computational techniques. Recently ANN has been developed as a powerful modelling tool in comparison to statistical or numerical methods [19]. It has been used in several applications including stock market prediction, speech recognition, manufacturing industries, food industries, chemical industries and biofuel industries to increase productivity [27]. ANN networks consist of input, hidden and output layers. Each neuron in ANN networks is properly connected to the other neurons. The number of neurons in the hidden layer or the number of hidden layers cannot be calculated. As a result, the number of neurons and hidden layers can be determined by trial and error methods [28].

In addition, the number of neurons and hidden layers were raised or decreased to bring the correlation coefficient closer to unity [26]. Babu et al. [23] optimized the parameters for improving biodiesel production from WCO using the RSM approach. It was noticed that the process parameters such as temperature, time, catalyst loading, molar ratio and stirrer speed influenced the biodiesel yield. Another study by Nahid Sultana et al. [29] predicted biodiesel production from microalgae using a Bayesian algorithm-based machine learning approach. This study

found that process parameters such as temperature, time, catalyst loading, molar ratio and stirrer speed affected the biodiesel yield. Many parameters influencing the biodiesel yield such as reaction temperature, time, molar ratio, catalyst loading and stirrer speed are chosen as input parameters. The input variables selection is based on the previous publication of Vinoth et al. [19], Mohammad et al. [30] and Akshay Garg et al. [31].

2.4.1. Choice of the network pattern

Babu et al. [18] stated that a combination of a backpropagation neural network and an LMNN training algorithm has accurately predicted the output. Neeta Kumari et al. [20] used three different networks such as BRNN, SCGNN and LMNN. Among the three networks, the BRNN network showed the most accurate prediction for reducing sugar yield. Another study by Vinoth Arul Raj et al. [21] used a feed-forward, MLP neural network and was analyzed by using the LM algorithm. The R^2 value for the ANN model was found to be 0.987, which showed that the model significantly fit with the experimental results. Sun et al. [32] employed three different networks such as KNN, DT and MLP networks to predict the biodiesel yield. The study revealed that MLP neural network along with the LM algorithm showed better prediction than any other models. Hence, in the present investigation, three different networks are used such as BRNN, SCGNN and LMNN, to predict biodiesel yield.

2.4.2. Model evaluation

Babu et al. [18] used statistical error parameters like regression coefficient (R^2), root mean square error (RMSE), mean absolute percentage error (MAPE), mean square relative error (MSRE) and Nash/Sutcliffe coefficient of efficiency (NSE) in their research work to evaluate the performance of the ANN proposed models. Ismail et al. [33] used error measurement standards in their study to evaluate the performance of the ANN models. The optimal measurement range should be within the range of $R > 0.90$, $MSE < 0.01$, and $MAPE < 5\%$. Vinoth Arul Raj et al. [21] suggested regression analysis for ANN and RSM models for evaluation. It found that the R^2 value of RSM and ANN was found to be 0.87 and 0.96 respectively. Vinoth et al. [19] implemented R^2 , MSE and RMSE error measurements for ANN model evaluation. The R^2 value of RSM and ANN was found to be 0.99, 0.0035 and 0.073 respectively. According to Sun et al. [32], ANN models performed well, with RMSE values of 0.49460, 0.18596, and 0.80422, respectively. The models also demonstrated good accuracy, with R^2 values higher than 0.9.

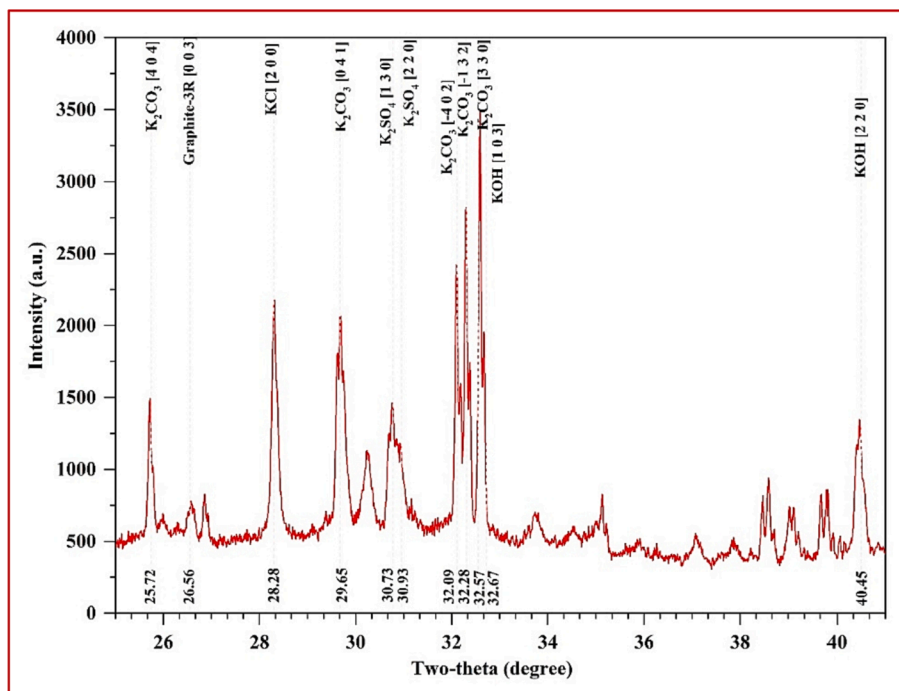


Fig. 3. X-ray diffraction of biocatalyst impregnated with KOH as a catalyst.

$$R^2 = 1 - \left(\frac{\sum_{i=1}^n (t_i - o_i)^2}{\sum_{i=1}^n (o_i)^2} \right) \quad (1)$$

$$RMSE = \left(\frac{1}{n} \sum_{i=1}^n (t_i - o_i)^2 \right) \quad (2)$$

$$MAPE = \frac{1}{n} \sum_{i=1}^n \left(\frac{t_i - o_i}{t_i} \right) \times 100 \quad (3)$$

RSM and ANN are the best statistical solutions in process modelling and successful tools for experimental modelling in process modelling. However, these models have not been applied to mixed cooking oil biodiesel synthesis using a biocatalyst impregnated with KOH as a catalyst. The ANN models are trained by using the experimental data obtained through a central composite design. The ANN architecture for the biodiesel synthesis model is shown in Fig. 2.

3. Results and discussion

3.1. Preparation of heterogeneous biocatalyst on biodiesel production from vegetable wastes

The biocatalyst was synthesized from vegetable wastes such as Chinese broccoli, Cauliflower and Napa cabbage according to the publication of Sumrit Mopoung et al. [34], and Vinoth et al. [19]. The collected vegetable samples were mixed, and their size was reduced to 1 cm by manual cutting. Furthermore, the size was decreased through grinding and selecting using an aluminium sieve. The mixed vegetable samples were aired outside until it was little vegetable surface moisture left. The dried samples were kept in an air force oven at 110 °C for 24 hr to remove the moisture once. The mixture samples were carbonized in tubular quartz at 800 °C for 3 hr, with a ramp rate of 5 °C/min, and under a nitrogen atmosphere. Since surface area did not significantly increase over 800 °C [19], hence this temperature was considered as optimal calcination temperature. The sample was then rinsed repeatedly with distilled water until the pH reached 6–7. In the end, activated carbons were obtained after the sample was kept in a hot air oven for 12

hr at 110 °C.

The potassium hydroxide (KOH) impregnated with a biocatalyst was synthesized by using the incipient wet impregnation method [19,23]. Different quantities of KOH (5, 10, 15 and 20 wt%) were impregnated on a biocatalyst based on the previous publication of Babu et al. [20]. The biodiesel yield increased from 73 % to 94 % when increased the KOH concentration from 5 wt% to 15 wt%. This is because of the intensification of basicity. Biodiesel production gets reduced when further increased the KOH concentration from 15 wt% to 20 wt%. This may be an increase in KOH concentration that promotes soap formation. Therefore, 15 wt% KOH was chosen for this experiment. Further, 15 wt % of KOH was loaded on 85 wt% in biocatalyst. The mixture was agitated for 24 hr at 32 °C after being liquefied in 30 ml of deionized water. The synthesized catalyzed was maintained at 110 °C in the muffle furnace for 6 h. Additionally, the catalyst was calcined in a muffle furnace at 530 °C for 5 h.

3.2. Heterogeneous biocatalyst and biodiesel characterization

3.2.1. Biocatalyst characterization through X-ray diffraction and SEM analysis

The X-ray diffraction pattern of the potassium-impregnated biocatalyst was captured by (Smartlab XE, Rigaku with D/teX Ultra 250) diffractometer, scan range $2\theta = 5\text{--}90^\circ$ with speed $10.00^\circ/\text{min}$, increasing step width 0.01° , using $\text{CuK}\alpha$ radiation ($\lambda = 1.542 \text{ \AA}$) and X-ray generator of 40 kV, 100 mA. Fig. 3. shows the wide-angle X-ray diffraction pattern of the KOH-impregnated biocatalyst. The characteristic peak of graphitic structure observed at $2\theta = 26.4^\circ$ corresponds to the (002) plane. The as-synthesized bio-catalyst primarily shows the reflection from the K_2CO_3 , KCl, K_2SO_4 and KOH, respectively. The peaks positioned at the 25.7° , 29.6° , 32.1° , 32.3° and 32.6° , corresponding to the (004), (041), (-402), (-132), and (330) planes of the face-centred crystal structure of K_2CO_3 . While the peaks at 29.9° and 30.04° correspond to the (130) and (220) plane I of face-centred crystal structure K_2SO_4 , respectively. The peaks around 32.7° and 40.4° confirm the existence of the KOH crystal structure. Although an additional peak at 28.28° reveals the (220) reflection plane of KCl. The average crystallized size of the KOH-impregnated biocatalyst was estimated to be

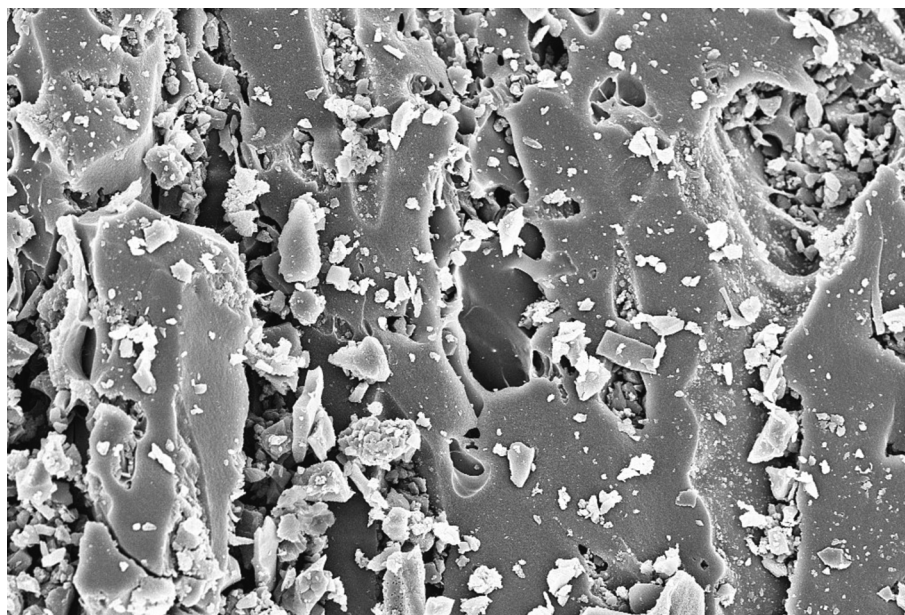


Fig.4. SEM image of KOH-impregnated biocatalyst on biodiesel production.

Table 2
GCMS analysis of mixed cooking oil methyl ester.

Peak No.	Ret. Time	% Area	Height%	A/H	Name
1	4.803	0.20	0.38	2.73	Dodecanoic acid, methyl ester
2	6.870	1.83	2.92	3.19	Methyl tetradecanoate ester
3	7.379	0.56	0.81	3.48	Methyl myristoleate ester
4	8.426	0.19	0.33	2.93	Pentadecanoic acid, methyl ester
5	10.559	74.75	68.21	5.58	Hexadecanoic acid, methyl ester
6	10.902	1.69	1.80	4.79	Methyl hexadecenoate
7	11.080	20.09	24.69	4.15	9-Hexadecenoic acid, methyl ester, (Z)-
8	11.965	0.30	0.40	3.85	<i>n</i> -Propyl 9,12-hexadecadienoate
9	13.135	0.25	0.29	4.36	Heptadecanoic acid, methyl ester
10	13.744	0.14	0.17	4.20	Methyl 8-heptadecenoate

56.67 nm by using the Debye Scherer formula. The morphology of the potassium hydroxide-impregnated biocatalyst is depicted in Fig. 4. The catalyst's morphology and lattice structure are measured by using scanning electron microscopy (Carl Zeiss Microscopy Ltd., UK). It was run at a voltage of 200 kV, a lattice resolution of 0.194 nm, and a point-to-point resolution of 0.14 nm. It was noted that the potassium hydroxide on the surface of the activated carbon is visible in the SEM image, suggesting that the KOH was preserved despite numerous water washes. Further, the potassium hydroxide was evenly distributed across the surface of the activated carbon indicating that KOH was uniformly deposited. The plate shape and average crystallite size of the particles were found to be 56.5 nm. There was no evidence of aggregation on the catalyst's surface.

3.2.2. Biodiesel characterization through gas chromatography and mass spectrometry

The fatty acid methyl ester composition was examined using Gas chromatography and mass spectrometry (GC–MS, QP 1000, Shimadzu Co., Kyoto Japan), it was fitted with a capillary column (DB-wax, 30 m × 0.25 mm, film 0.25 μm). Fatty acid compounds were found in GC–MS analysis using a flame ionization detector. Table 2 demonstrates the GC–MS analysis of mixed waste cooking oil methyl ester. The sample was inoculated into the injector while the furnace was kept at a constant temperature of 280 °C. Helium gas was fed into GC–MS at a flow rate of 1 ml/minute. From a beginning temperature of 100 °C to a final temperature of 280 °C, the oven was heated. The run was completed in 14

min and the collected data was saved in the library. GC–MS spectrum of FAME is depicted in Fig. 5, and Table 2 lists the fatty acid composition of MWCO. From Fig. 5, it can be seen that the Methyl tetradecanoate, Hexadecanoic acid methyl ester and 9-Hexadecenoic acid methyl ester were found at 6.78 min, 10.55 min and 11 min respectively. Vegetable and waste cooking oil species name and oil content is given in Table 3.

3.3. Analysis of variance (ANOVA) for central composite design model

The significance of the model was evaluated by using lack of fit, analysis of variance, and coefficient of determination (R^2). The ANOVA result for the biodiesel model is given in Table 4. The quadratic model was significant as the p-value was <0.05. Additionally, the P-value in Lack-of-fit was <0.05 reporting the assurance of the model. The significant process parameters were observed from the ANOVA (Table 4). Furthermore, the R^2 and adjusted R^2 values were found to be 0.983, and 0.981, respectively. The reliability of the anticipated model is indicated by an R^2 value greater than 0.98 reported in earlier studies [11,13,23]. Overall, R^2 , Lack of Fit and P-value were attained from the investigation that supported the assurance of the model. Following the study of model significance, each term's significance shows how various factors and their interactions affect the synthesis of biodiesel using a biocatalyst impregnated with KOH as a catalyst.

The accuracy of the model is designated by the coefficient of determination (R^2) and adjusted R^2 which determine the number of predictors in the model. A point prediction method was used to measure the

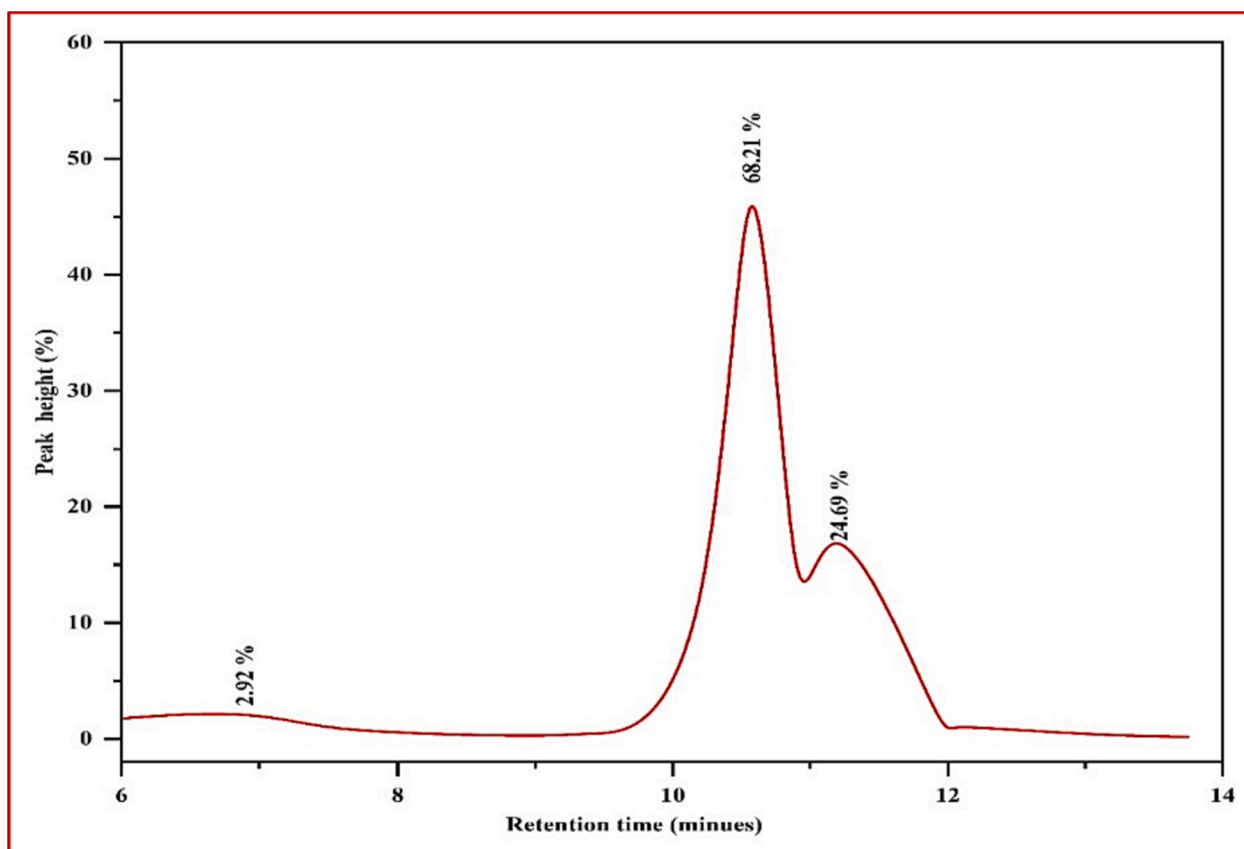


Fig. 5. GCMS spectrum of mixed cooking oil methyl ester.

Table 3

Vegetable and waste cooking oil species name and oil content [35].

Type of oil	Common name	Species name	Oil content (wt.%)
Vegetable oil	Sunflower	<i>Helianthus annuus</i>	25–35
	Palm	<i>Elaeis guineensis</i>	30–60
	Soybean	<i>Glycine max</i>	18–20
	Rice bran	<i>Oryza sativa</i>	15–23
	Rapeseed	<i>Brassica napus</i>	35–46
Waste cooking oil	Sunflower and Palm oil	<i>Helianthus annuus</i> and <i>Elaeis guineensis</i>	25–40

optimal conditions in biodiesel synthesis. The optimum biodiesel yield of 94.5 % was achieved for the biocatalyst impregnated with KOH at a reaction temperature of 55 °C, a molar ratio of 1:9, and a catalyst loading of 7 wt% and 120 min of reaction time and stirrer speed of 900 rpm. The experiments were repeated five times to check the viability of predicted values and the yield was found to be 94.7 %. The second-order polynomial equation expressed the relationship between independent variables and response.

3.4. Interaction effect of process parameters on biodiesel yield through RSM approach

3.4.1. Effect of temperature and molar ratio on biodiesel yield

Fig. 6a illustrates the interaction effect of molar ratio and temperature on biodiesel yield. The reaction temperature and molar ratio are the key factors, which are influences the biodiesel yield. From Fig. 6a, it was shown that as the temperature rose from 45 °C to 60 °C, the biodiesel yield increased. This may be due to a rise in temperature promoting the collisions between the reactants, and improving the mass transfer between methanol and oil, thus increasing the conversion rate. Whereas, no improvement was found in biodiesel yield when further increased the reaction temperature from 60 °C to 65 °C. This may be due to methanol starting to vaporize above 60 °C and the system becoming two phases thereby reducing the conversion rate. Similarly, the biodiesel yield increased with an increase in a molar ratio from 1:3 to 1:7 up to 94 %, whereas the yield gets reduced from 94 to 73 % when further increasing the molar ratio from 1:7 to 1:9. This is because too much methanol makes glycerol more soluble, which improves the emulsification between glycerol and biodiesel. As a result of the reversible reaction, the biodiesel yield got reduced. However, the mass transfer between biocatalyst and liquid is limited by the larger molar ratio and higher tem-

$$Y = +57.34 - 4.56A + 11.34B + 1.54C + 22.07D - 0.97E + 4.37AC + 6.54AD - 7.61BC - 6.12BD + 5.60BE - 1.67CD - 11.64CE + 5.57ED + 6.54A^2 + 3.32B^2 - 8.90C^2 + 5.32AD^2 - 0.43E^2 \quad (4)$$

Table 4
ANOVA results for the biodiesel model.

Source	Sum of Squares	df	Mean Square	F-Value	Prob > F	
Model	2170.45	20	108.52	41.33	< 0.0001	Significant
A-Molar ratio	275.4	1	275.4	17.45	0.0024	
B-Catalyst concentration	252.67	1	252.67	16.52	0.0035	
C-Reaction Temperature	112.12	1	112.12	6.88	0.0246	Significant
D-Reaction Time	12	1	12	782.14	<0.0001	
E-Stirrer Speed	132.43	1	132.43	8.31	0.0456	Significant
AB	120.21	1	120.21	7.42	0.017	Significant
AC	21.47	1	21.47	1.34	0.2763	Insignificant
AD	36.14	1	36.14	2.25	0.0176	Significant
AE	706.30	1	706.30	44.34	<0.0001	
BC	397.45	1	397.45	25.42	0.0003	
BD	11.08	1	11.08	0.75	0.4014	Insignificant
BE	561.10	1	561.10	33.44	<0.0001	
CD	95.41	1	95.41	6.81	0.0344	
CE	554.12	1	554.12	35.02	0.001	
DE	267.14	1	267.14	16.45	0.0016	
A ²	1.98	1	1.98	0.12	0.789	Insignificant
B ²	124.34	1	125.65	7.70	0.0171	
C ²	40.61	1	40.61	2.49	0.1567	Insignificant
D ²	3222.6	1	3222.6	197.45	<0.0001	
E ²	164.08	1	164.08	10.05	0.0078	
Residual	178.11	10	178.11			
Lack of Fit	11.45	11	11.45	1.45	0.567	Insignificant

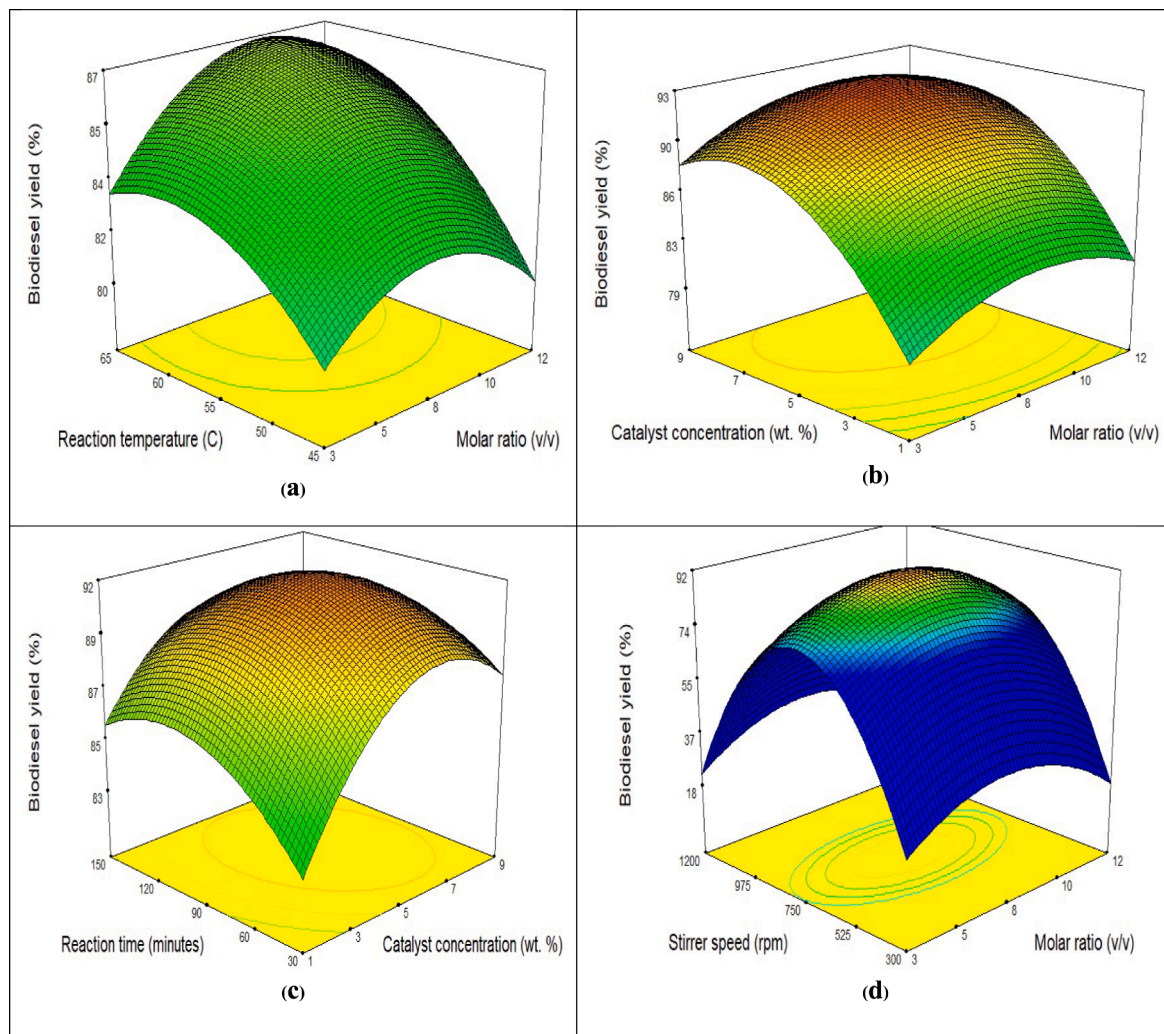


Fig. 6. Interaction effect of (a) temperature and molar ratio (b) molar ratio and catalyst concentration (c) reaction time and catalyst loading (d) stirrer speed and molar ratio on biodiesel yield.

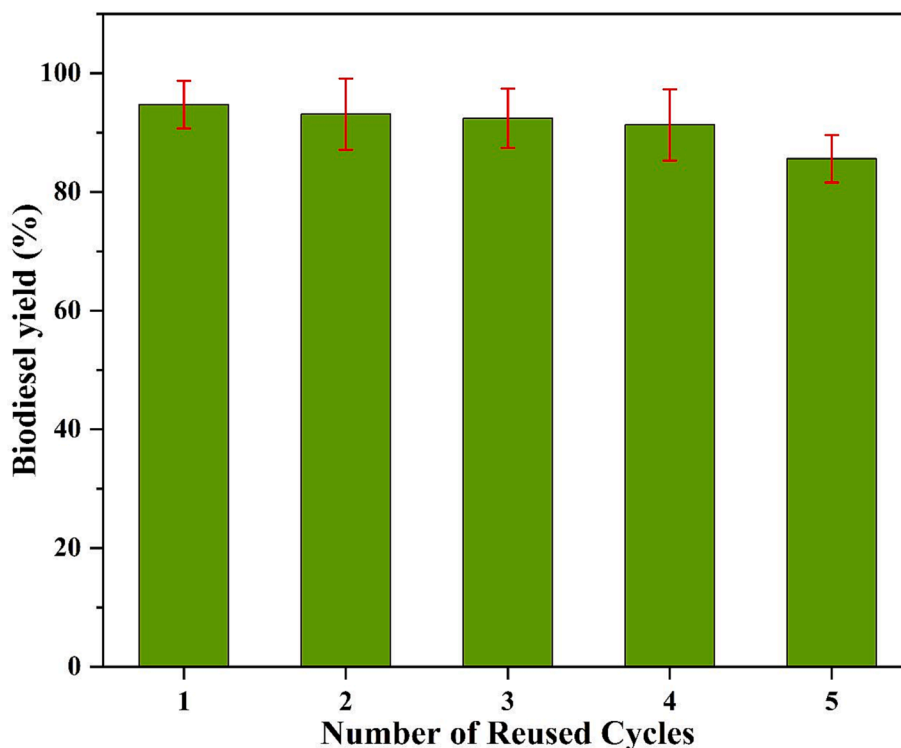


Fig. 7. Reusability of biocatalyst on biodiesel synthesis.

perature, resulting in a lower biodiesel yield as observed by Vinoth et al. [19]. In addition, the minimum yield was observed at a lesser temperature and molar ratio because of the less methanol content and low reaction rate. The optimum biodiesel yield of 94.7 % was found at a 1:7 M ratio, which was higher than the earlier published work by Babu et al. [23] achieved a yield of 93 % at a 1:12 M ratio and a reaction temperature of 55 °C. Iyyappan et al. [36] synthesized biodiesel from *Caulerpa racemosa* utilizing a recombinant *pichia pastoris* cell biocatalyst. The maximum biodiesel yield of 93.64 % was achieved at 30hr of reaction duration and a temperature of 45°C. Vinoth et al. [19] produced biodiesel from environmentally beneficial and non-edible *Aegle Marmelos* Correa seed oil using a biocatalyst. By undergoing microwave transesterification for 20 min at 55 °C with a 1:12 M ratio, the 4.5 wt% catalysts have achieved the optimum biodiesel yield of 92 %.

3.4.2. Interaction effect of molar ratio and catalyst concentration on biodiesel yield

Fig. 6b depicts the effect of catalyst loading and molar ratio on methyl ester yield. The surface plot revealed that the biodiesel yield increases as catalyst loading rises from 1 wt% to 7 wt% and molar ratio 1:3 to 1:7 respectively. At a catalyst loading of 7 wt% and a molar ratio of 1:7, the highest yield of 94.7 % was attained. A similar observation was noticed in the previously published work by [37,8]. Niranj Daimary et al. [37] achieved the maximum biodiesel yield of 97 % at a catalyst loading of 4.5 wt% and a molar ratio of 1:9 and a reaction temperature of 55 °C. Zulfiqar et al. [8] achieved a maximum biodiesel yield of 92 % at a catalyst loading of 10 wt% and 1:6 M ratio. Another study by Arumugam et al. [25] extracted a biodiesel yield of around 93 % at a molar ratio of 1:8 and a catalyst loading of 15 wt%. More methanol and oil ratio is needed to achieve the maximum biodiesel yield because water concentration may lower biodiesel production. When the catalyst loading rises above 7 wt% and a molar ratio rises above 1:9, the biodiesel production decreases due to high mass transfer brought on by the higher catalyst concentration causing the mixture to become more viscous, which lowers the yield [23]. Moreover, higher catalyst loading causes soap formation. Further, excessive methanol reverses the

transesterification reaction by deactivating the catalyst as observed by Srilatha et al. [38]. Also, it was discovered that in the absence of a catalyst no yield was seen. Che Zhao et al. [39] synthesized a highly effective and reliable CaO-based catalyst that was used to catalyze the transesterification of vegetable oil with methanol using rice husk biochar as a catalyst. The outcomes showed that the highest biodiesel yield of 93.4 % was achieved at 30 % CaO catalyst loading and 700 °C.

3.4.3. The influence of reaction time and catalyst concentration on biodiesel yield.

Fig. 6c shows the surface plot of time and catalyst concentration on biodiesel yield. According to the plot, the biodiesel yield increases as the reaction time from 30 min to 120 min and decreases when it further increases the reaction time from 120 min to 150 min. This may be due to the high viscosity of the mixture makes it difficult for biodiesel separation. Biodiesel harvest augmented with a rise in catalyst loading up to 7 wt% and no improvement was found in biodiesel yield when further increases the catalyst concentration was as can be observed from Fig. 6c. More amount of catalyst present in transesterification reaction causes soap formation resulting in lower yield. According to the experimental data, the maximum biodiesel yield of 94.7 % was attained at a reaction duration of 120 min and catalyst loading of 7 wt%, which was higher than the prior published works by Takase et al. [40] and Sneha et al. [6]. The highest biodiesel yield of 90.8 % was observed by Takase et al. [40] and 83.6 % was achieved by Sneha et al. [6]. Biodiesel production was minimum at a lower reaction duration and catalyst loading may be due to poor mixture and reaction rate. Further, the biodiesel yield decreases when increased the reaction time and catalyst loading due to the saponification reaction. In another study, Jiayan Wang et al. [41] developed a new solid base catalyst from gasified straw slag with high stability for efficient biodiesel production, with a 20 % catalyst and a molar ratio of 1:12 and good catalytic activity and stability converting fatty acid to methyl ester 90 % at a temperature of 200 °C for 8 hr of reaction duration.

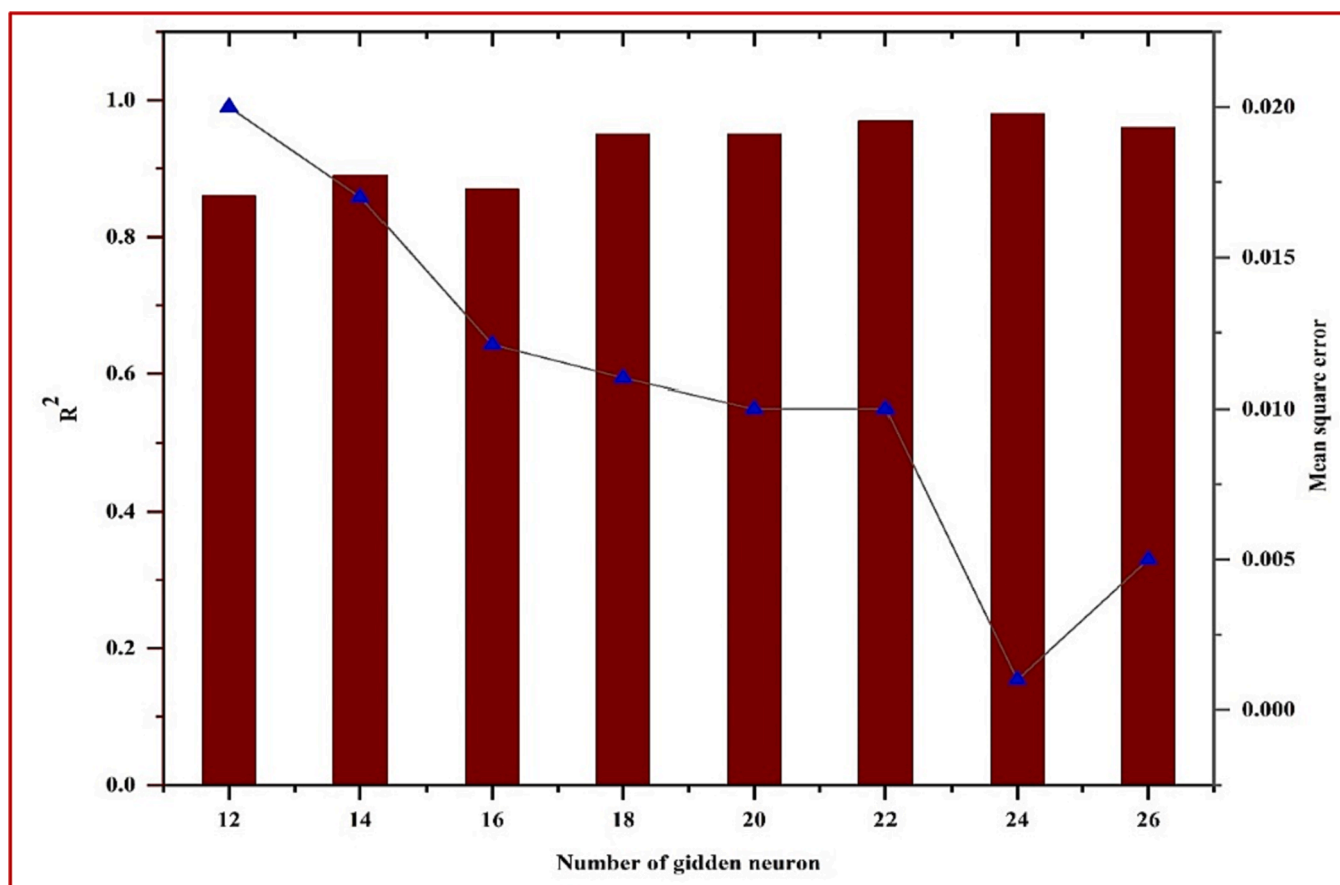


Fig. 8. Selection of numbers of neurons for biodiesel model based on R^2 and RME.

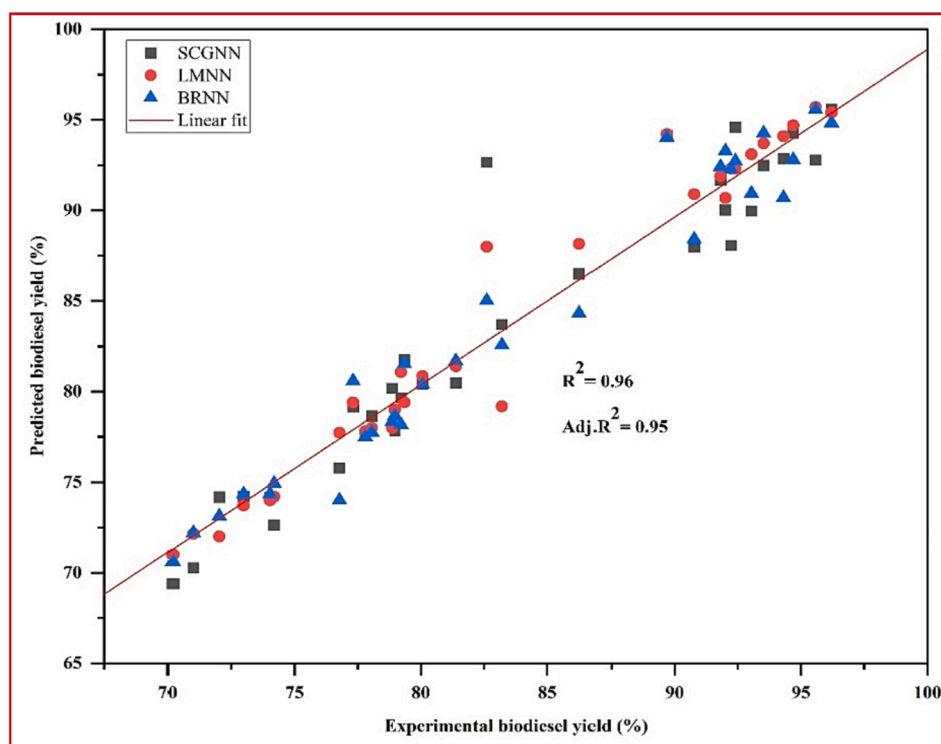


Fig. 9. Predicted results of ANN model vs RSM model.

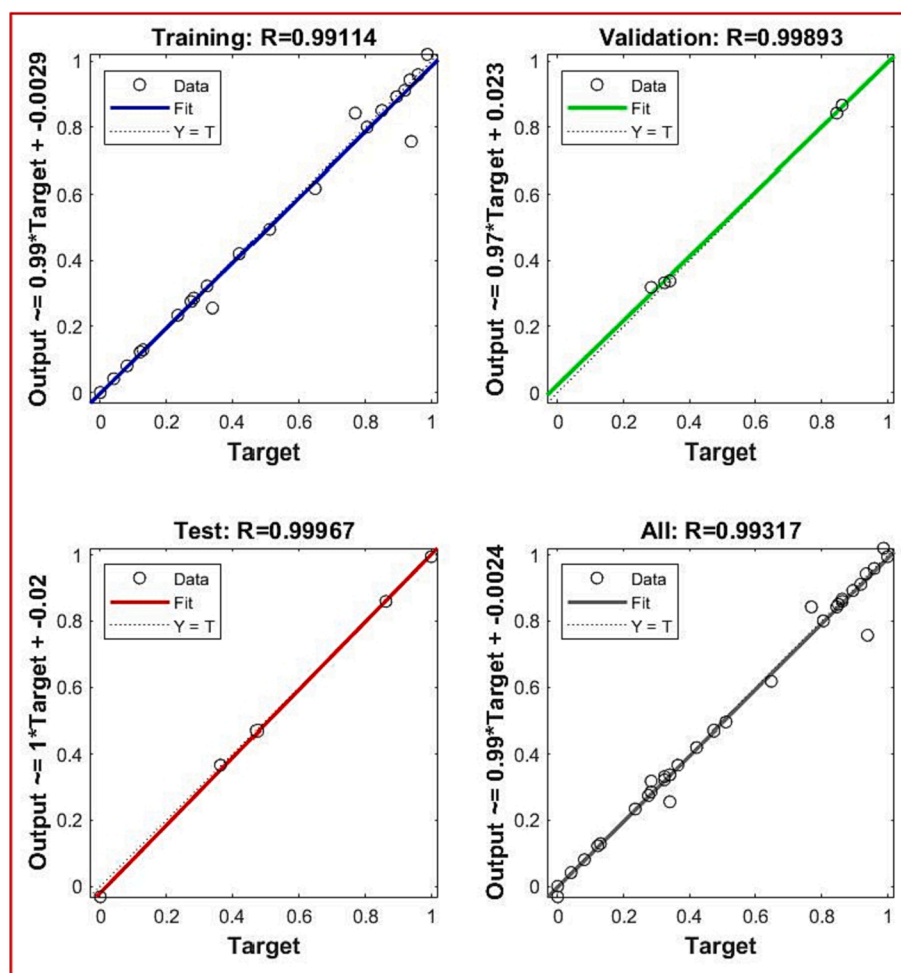


Fig. 10. Overall correlation coefficient for ANN biodiesel model.

3.4.4. The influence of stirrer speed and molar ratio on biodiesel yield

Fig. 6d demonstrates the interaction effect of stirrer speed and molar ratio on biodiesel production. From the surface plot, it was perceived that the biodiesel production was improved with an intensification of stirrer speed and molar ratio up to 900 rpm and 1:9 respectively. The biodiesel yield deteriorated when increases the stirrer speed and molar ratio were beyond 900 rpm and 1:9 M ratio. This may be due to the high stirrer speed making the mixture too viscous resulting in the lowest biodiesel yield, creating difficulty in biodiesel separation from glycerol. Further, no improvement was noticed in biodiesel yield with a molar ratio increased beyond 1:9 due to the possibility of reverse transesterification reaction which leads to dropped biodiesel yield. In addition, at a lower stirrer speed and molar ratio, the minimum yield was observed. This is because of the improper mixture of methanol and cooking oil, also a low amount of methanol reduces the transesterification reaction rate. Additionally, the yield decreased as the molar ratio rose above 1:12. This is because too much methanol makes glycerol more soluble, which improves the emulsification of glycerol and biodiesel. As a result of the reversible reaction, methyl ester production gets reduced. However, the larger molar ratio and higher temperature, resulted in a lower biodiesel yield as noticed by Srilatha et al. [38]. The reusability test was carried out for a biocatalyst impregnated with KOH as a catalyst at an optimal reaction condition at a molar ratio of 1:9, and catalyst concentration of 7.4 wt%, reaction temperature of 56 °C and stirrer speed of 912 rpm. From Fig. 7, it can be observed that the first cycle of the reaction produced a 93.4 % yield of biodiesel. After the reaction, the used catalyst was removed from the reactor, dried in a hot air oven and calcinated at 530 °C for further processing. However, in

the second cycle of the reaction and again in the fifth cycle, the biodiesel yield reduced to 91.3 % and 85.6 % respectively due to potassium ions leaching, instability of particles and lower reaction rate. From a sustainable and economical perspective, it is particularly beneficial to employ catalysts made from biomass sources. This is because of their excellent thermal and mechanical stability. Further, carbon or activated carbons reduced the cost of biodiesel production.

3.5. Comparison of experimental results vs ANN predicted values

The artificial neural network was employed in this work to predict the biodiesel yield using a biocatalyst impregnated with KOH as a catalyst, molar ratio, reaction time, temperature and stirrer speed as input variables. As a result of the nonlinearity of the transfer function, neural networks are more effective than linear transformations and performance functions are optimized by mean square error. For each iteration, the number of neurons varied from 12 to 26 and the topology research employed the MSE as a criterion. The data shown in Table 1 were used to construct and train the neural network. A total of 32 data points are used to construct the ANN model. Among these, 70 % of the experimental data was used to train the network and 15 % of the data could be used to test the network, the remaining 15 % of the data was used to validate the ANN model. Both input and output values are normalized before the training session. The purpose of normalization is to avoid overflow situations. When the network error (MSE) falls to a low value ($MSE \leq E_o$), the network training must end. In the current study, the training was completed after 9 iterations (epochs). The optimal feed-forward network model for the neural network is depicted

in Fig. 8. Five neurons in the input layer, twenty-four neurons in the hidden layer, and one neuron in the output layer were found to be the optimal design for the biodiesel model (5–24–1). According to Kshirsagar et al. [26], the best design for the emission model was determined to be (4–16–6), while the best architecture for the performance model was (4–16–3). Similarly, Babu et al. [18] found the optimal architecture for the performance model to be (4–13–5).

The degree of agreement between experimental and model predicted values is shown in Fig. 9. the strong correlation coefficient ($R^2 = 0.997$) and adjusted correlation coefficient ($R^2 = 0.987$) of the produced ANN model demonstrate its significance and suggest that it can be used to forecast the optimum topology. The majority of the experimental results are shown on the 45-degree line in Fig. 9, demonstrating how well the expected outcomes match with the real data. The potential of the ANN model can be improved by using normalization, target data into training, validation and test data subsets. According to Fig. 9, the R and RMSE values of the biodiesel model fall between 0.97 and 0.986 and 0.0123 and 0.0166 respectively. As a result, the ANN model may be effectively used to estimate and predict the biodiesel yield from mixed waste cooking oil pretreated with phosphoric acid. A similar finding was observed in published work [21,42]. Vinoth raj et al. [21] observed the R and RMSE values for the biodiesel model in the range of 0.97–0.986, and 0.0123–0.0166, respectively. Similarly, Vinoj et al. [42] noticed that for all the ANN models, the predicted values were highly associated with experimental values ($R^2 = 0.97$), showing higher prediction accuracy than RSM.

The correlation coefficient of the network for the phosphoric acid pretreated biodiesel model is shown in Fig. 10. The R-value in training, testing, and validation was 0.99, 0.99 and 0.99 respectively, as well as the overall R-value was 0.99 as can be shown in Fig. 10. A similar result was found from the earlier research studies. Leucaena leucocephala pretreatment was optimized using an ANN approach by Neeta Kumari et al. [20] for bioethanol production. According to the study, a superior prediction is made by the LMNN model, which had MSE and R values of 0.07 and 0.99 respectively. Similarly, Babu et al. [18] optimized the engine parameters using an ANN approach. The R-value of testing, training and validation was found to be 0.99, 0.99 and 0.98 respectively, with an overall R-value of 0.99. In another study, Kannan et al. [27] found similar kinds of R-values for the biodiesel model such as 0.97, 0.98 and 0.98 respectively for training, testing and validation with an overall correlation coefficient of 0.99. Hence, the above R-values show the ANN prediction accuracy and reliability of the ANN model compared to RSM.

4. Conclusion

The effect of biocatalysts impregnated with KOH on biodiesel production from mixed waste cooking oil was investigated experimentally. Optimization tools such as response surface methodology and artificial neural network were employed to optimize the process parameters in biodiesel production to obtain maximum yield with minimum methanol and catalyst, energy consumption. The main conclusions drawn from the experimental and predicted results are summarized as follows. The maximum biodiesel yield of 94.7 % was attained at a molar ratio of 1:9, and catalyst loading of 7 wt%, reaction temperature of 60 °C and stirrer speed of 900 rpm. Whereas, the optimum yield of 93.4 % was attained at a molar ratio of 1:9, with catalyst loading of 7.4 wt%, reaction temperature of 56 °C and stirrer speed of 912 rpm to be found by the RSM approach. A heterogeneous catalyst has the advantage of being free from separation, and purification problems over a homogeneous catalyst, and has a higher reactivity, large surface area and high porosity. Bayesian Regularization Neural Network and Transigmoid activation function were found to be the best algorithm in biodiesel process optimization when compared to the other optimization techniques. The optimal architecture for the biodiesel model was (5–24–1). The ANN and RSM models had lower R^2 , RMSE, and MAPE values, indicating the excellent

dependability of these two approaches for process optimization in biodiesel production. The current research could be expanded to examine biodiesel fuel's spray properties and particle size distribution. Additionally, computational fluid dynamics may be used to simulate the burning of biodiesel in a diesel engine. Hence, this research concludes that mixed waste cooking oil-based biodiesel could be a promising substitute for conventional diesel fuel in diesel engines.

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.

Data availability

Data will be made available on request.

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