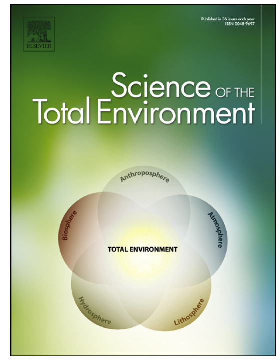

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Abstract

Transesterification is a promising technology for the biodiesel production to provide an alternative fuel that considers the environmental concerns. From the economic and environmental protection points of view, utilization of waste frying oil for the production of biodiesel addresses very beneficial impacts. Production of higher yield of biodiesel is a challenging process in order to commercialize it with a lower cost. The current study focuses on the influence of different parameters such as reaction temperature (ºC), reaction period (min), oil to methanol ratio and amount of catalyst (wt%) on the production of biodiesel. The main objective of this work is to develop a model via fuzzy logic approach in order to maximize the biodiesel produced from waste frying oil using montmorillonite Clay K-30 as a catalyst. The optimization for the operating parameters has been performed via particle swarm optimization (PSO) approach. During the optimization process, the decision variables were represented by four different operating parameters: temperature (40-140ºC), reaction period (60-300 min), oil/methanol ratio (1:6-1:18) and amount of catalyst (1-5 wt%). The model has been validated with the experimental data and compared with the optimal results reported based on other optimization techniques. Results showed the increment of biodiesel production by 15% using the proposed strategy compared to the earlier study. The obtained biodiesel production yield reached 93.70% with the optimal parameters for a temperature at 69.66ºC, a reaction period of 300 min, oil/methanol ratio of 1:9 and an amount of catalyst of 5 wt%.

Keywords: waste frying oil; biodiesel; optimization; fuzzy logic; clay-based catalyst

1. Introduction

Nowadays fossil fuel is still occupying the position of the main source of energy all over the world. But unfortunately, it is limited, depleting and has many negative environmental impacts (Brevik et al., 2019). Renewable and alternative energy sources are environment friendly compared to the traditional fossil fuel (Aransiola et al., 2014; Sayed et al., 2019). Biomass is a promising energy resource because it is renewable as well as abundant. (Hwangbo et al., 2019; Rafael et al., 2015; Abdelkareem et al., 2019). Biodiesel obtained from biomass can be an alternative to the conventional diesel and carbon neutral fuels (Fawaz and Salam, 2018, Wang et al., 2018). The main challenge facing the commercialization of the biomass-based biodiesel is its high cost which is significantly dependent on its biomass (Santori et al., 2012). Waste frying oil is a hazardous material with harmful impacts on the environment and it is worthy if it can be well treated and converted to a useful product. Waste frying oil is considered not only as a cost effective raw material for producing biodiesel but also with a competitive cost to that obtained from fossil fuel (Uzun et al., 2012).

Catalysts play an important role in the biodiesel production process. Heterogeneous catalysts such as alkaline earth oxides, zeolites, inorganic-oxide solid acids hydrotalcites, and ion-exchange resins are usually used for biodiesel production (Abdullah et al., 2017). Compared to the above catalysts, clay is one of the cost effective catalysts due to its high activity in heterogeneous reactions particularly the transesterification ones (Alves et al., 2014). Different studies have been carried out to investigate the effect of different parameters of the transesterification process from oil into biodiesel, e.g., reaction time, temperature, solvent to oil ratio, and amount of catalyst (Ashok et al., 2018; de Araújo et al., 2013; Hamze et al., 2015; Jaimasith and Phiyanalinmat, 2007; Nguyen et al., 2018; Peng et al., 2018; Said et al., 2015).

Using composite rotatable design and Response Surface Methodology (RSM), Chin et al., (2009) investigated the optimal operating parameters affecting the biodiesel produced from waste palm cooking oil using oil palm as a catalyst. The experimental results revealed a biodiesel production of 71.74% at optimum operating conditions. Using microwave heating system, Peng et al., (2018) demonstrated that eggshell is a catalyst which can be utilized effectively to obtain biodiesel production from waste cooking oil. A biodiesel yield of 87.8% was achieved at optimum operating conditions. In another study, Ashok et al. (2018) demonstrated that MgO is a catalyst that can effectively produce biodiesel from waste cooking oil. A high biodiesel yield of 93.3 % was achieved at the optimum operating conditions of methanol to oil ratio, reaction time and wt% of catalyst. Moreover, the authors demonstrated that the MgO catalyst was effectively used for five times without a noticeable decrease in biodiesel yield. Due to the several advantages of static mixture reactor such as no moving parts, ease in handling viscous solutions, Nguyen et al. (2018) applied static mixer reactor for producing biodiesel from waste cooking oil for the first time. Using a quadratic model and applying the RSM optimization method, the static mixer reactor revealed a very high oil yield that reached 97.8% at the optimum operating conditions. Using RSM, Hamze et al. (2015) reported that biodiesel produced from waste cooking oil (Soybean and sunflower oil) could be as high as 99.38% using optimal experimental operating conditions of methanol to oil of 7.5 to 1, KOH catalyst (1.4 wt%), reaction time of 60 minutes at 65^{o}C. The optimization process is very close to that of the experimental one, where its prediction reached 99.5%. Due to the demonstrated and promising results of using clay for biodiesel production, Jaimasith and Phiyanalinmat (2007) selected it as a catalyst. In a recent study, Ayoub et al., (2016) have demonstrated that montmorillonite clay K-30 is an effective catalyst for the biodiesel production from waste frying oil. Using response surface methodology, a biodiesel production rate of 78.4% was obtained at an optimized operating condition. The aim of the current research work is to develop a fuzzy model based on swarm optimization to predict the optimum operating conditions that maximize the amount of biodiesel production. Furthermore, the developed model could be applied for the commercial applications.

2. Materials and Methods

2.1. Fuzzy Modelling for Biodiesel System

Boolean logic (BL) was the only algebraic logic known in the era before 1965 when Zadeh introduced the concept of fuzzy logic (FL). BL is based on only binary valued set, {0, 1}, however, FL is a more generalized logic which is based on multi-valued set. Moreover, FL is mimicking the human-like description of event occurrence. Therefore, in data modeling, FL is an efficient tool to build a model from uncertain and/or noisy data. The procedure of fuzzy modeling includes three sequential steps begins by the fuzzification process and ends by the defuzzification process and in between is the inference system. With the help of membership functions (MFs), converting values from crisp to fuzzy and vice versa is what is meant by fuzzification and defuzzification processes, respectively. The main part of the inference system is the rule-base which in this case study is generated from the data.

The *IF* (Inputs) *THEN* (Output) forms the rule's structure. The fuzzy rule is constructed by either Mamdani or Takagi-Sugeno-Kang (TSK) types. The rule's output in Mamdani is formulated by a fuzzy MF while in TSK it is formulated by either a linear or nonlinear relationship between the inputs and the output. The Center of Gravity (COG) defuzzification method is popular in Mamdani-type while the Weighted Average (Wtaver) is the suitable one in TSK-type.

To enhance the production of biodiesel from waste frying oil over Montmorillonite Clay K-30, some operating variables, which have great influence on the system's performance, are considered. These variables are the reaction time (minutes), reaction temperature (°C), amount of catalyst (wt%) and oil/methanol ratio. Ayoub et al. (2016) studied the effect of these four controlling variables on the biodiesel production in 30 experimental runs. Commercial clay catalyst montmorillonite K-30 (in powdered form) has been utilized in the experimental work because it has several advantages such as safe to handle, reusable, inexpensive and can act as a general acid or base. Furthermore, Montmorillonite K-30 clay catalyst also considered as environmentally safe. In addition, such catalyst can be recycled which increases the economic efficiency of the overall production process.

In Ayoub et al., every individual experiment was run with a different combination and different values of the controlling variables. The authors recorded the resulting input-output data and then used the statistical tools to analyze and build a model for the biodiesel production system. With a deep investigation of the plots of the experimental data of the biodiesel system, it can be noticed that the data represents a highly nonlinear performance and might contain uncertain data because of noise. Consequently, the fuzzy modeling procedure is the most suitable choice due to its efficiency in capturing as well as tracking these kinds of input-output relationships.

In the current work, a fuzzy model is constructed based on the 30 experimental runs with the 4-input and one-output dataset conducted by Ayoub et al. (2016). During the optimization process, four operating parameters of biodiesel production process; temperature, reaction time, oil/methanol ratio and amount of catalyst were used as the decision variables. However, with this small number of experimental dataset, the data is divided into two subsets one for training and the other for testing with a ratio of 80 to 20, respectively. Therefore, the number of training and testing data points are 24 and 6, respectively.

In fuzzy modeling, the structure and rule-base of the model have to be configured primarily before the training process starts. Therefore, a TSK-type ANFIS comprises 4-input and one linear output is built and the Subtractive Clustering (SC) method is used to configure the rule-base. The number of rules was 21 rules. The Gaussian-shape MF and Wtaver are used for fuzzification and defuzzification methods, respectively. The system's training process used the 24 records of the training subset and applied them for 30 epochs. To guarantee an accurate training phase, the model is trained until a satisfying small testing error is reached. Also, the mean squared errors, MSE, between the model outputs and the experimental data, were computed for both training and testing phases. The MSEs were found to be 14.8558 and 4.6593 for training and testing data, respectively.

2.2. Particle Swarm Optimization (PSO) for Biodiesel System

Some optimizers are imitated from the natural social behavior and dynamic movements with communications of some creatures such as ants, bees, birds, etc. PSO is a similar algorithm that formulates the bird's movements mathematically. In 1995, Kennedy and Eberhart succeeded to present the mathematical formulation of PSO to be used in the optimization problems. The main and basic idea of the PSO algorithm is in the deployment of a number of agents (particles) that constitute a swarm moving around in the search space looking for the best solution (position). Each particle in the search space adjusts its "flying" direction and position according to its own flying experience as well as the flying experience of other particles in the swarm. The movement is expected to be attracted towards a promising area to reach the global optimum. Every particle in the swarm preserves to track its best position, personal best, *pbest* as well as the best position of the whole particles, global best, *gbest*. The updating rule of the particle's location is illustrated in Eqs 1 and 2.

$$p\,(k+1) = p(k) + v\,(k+1) \tag{1}$$

$$v\,(k+1) = w * v\,(k) + c_1 * r_1 * (pBest - p) + c_2 * r_2 * (gBest - p) \tag{2}$$

where, p is the particle's position; v is the path direction; $c_1$is the weight of local experience; c_2 is the global experience; *pBest* is the particle's best position of; *gBest* is the best position of all particles encountered throughout the iterations; r_1 and r_2 are random variables changing from 0 to 1.

In this work, the optimization problem is stated as "what are the optimal values of reaction time (minutes), reaction temperature (°C), amount of catalyst (wt%) and oil/methanol ratio so as to maximize the biodiesel production yields' percentage?". In the optimization process, the problem can be converted from maximization to minimization and vice versa by multiplying the objective function with -1. Hence, the objective function will be:

$$f(x) = -(Yields)$$

where, *Yields* is the percentage of biodiesel production yields and x is the decision variables vector. The optimization process configuration of the biodiesel system's, applied in the current study, is presented in Fig. 1. The optimization's controlling parameters are defined with a maximum iterations of 30, a population size of 30, a local experience of 1.5, a global experience of 2.0, and a weight of inertia of 1. The lower and upper bounds of the reaction time, reaction temperature, amount of catalyst and oil/methanol ratio are taken as: [60 300] (minutes), [40 140] (Co), [1 5] (wt %), [1/6 1/18], respectively.

To confirm the stability of the results and to be away from getting results by randomness, a statistical evaluation is done. These statistical metrics include the minimum (Min), maximum (Max), average (Avg), and standard deviation (StD) of the obtained data.

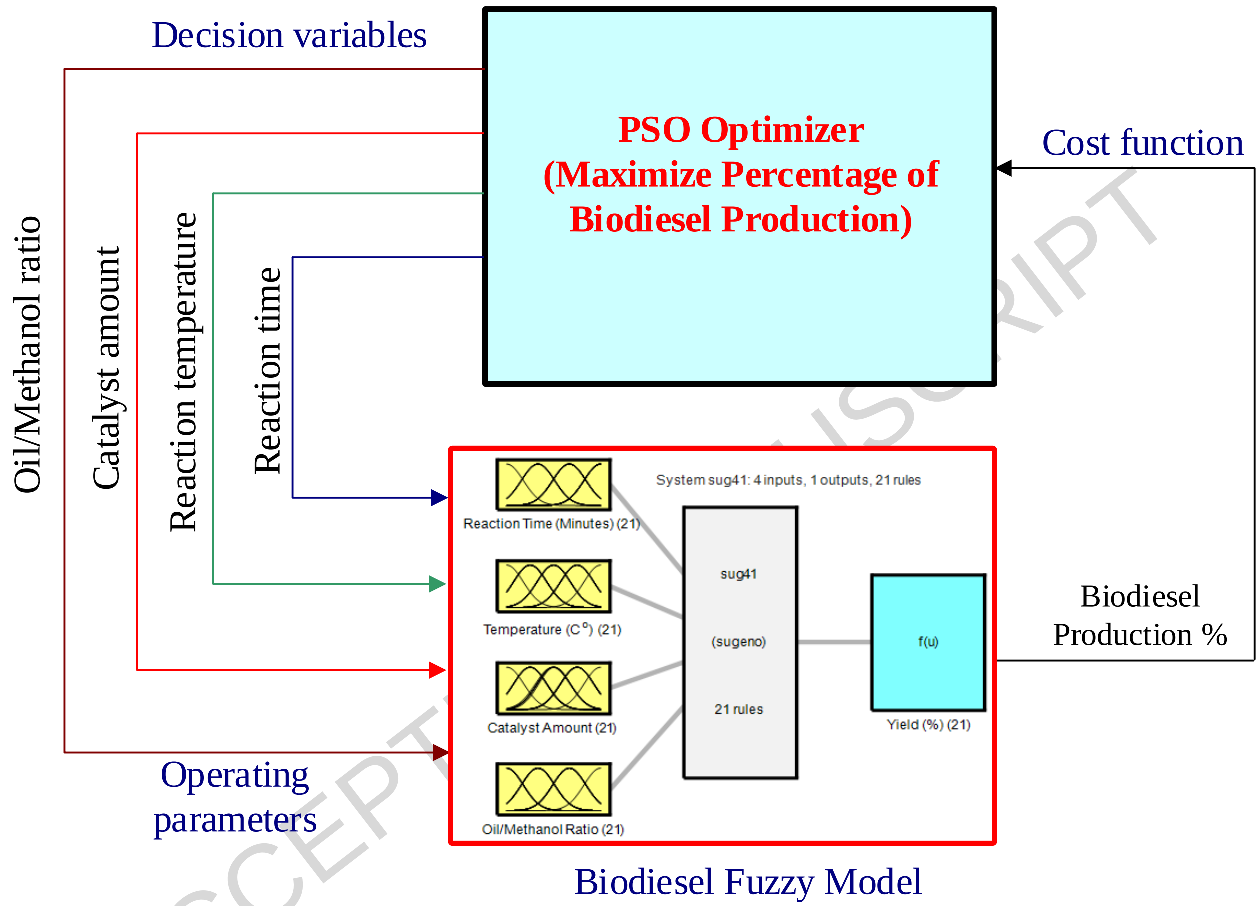

Fig. 1. The configuration of the biodiesel system's optimization process

3. Results and Discussion

During the training phase, the best model structure is considered when the testing MSE was the lowest among all training trials. Fig. 2 shows the plots for both training and testing phases of the fuzzy model predictions against the experimental data.

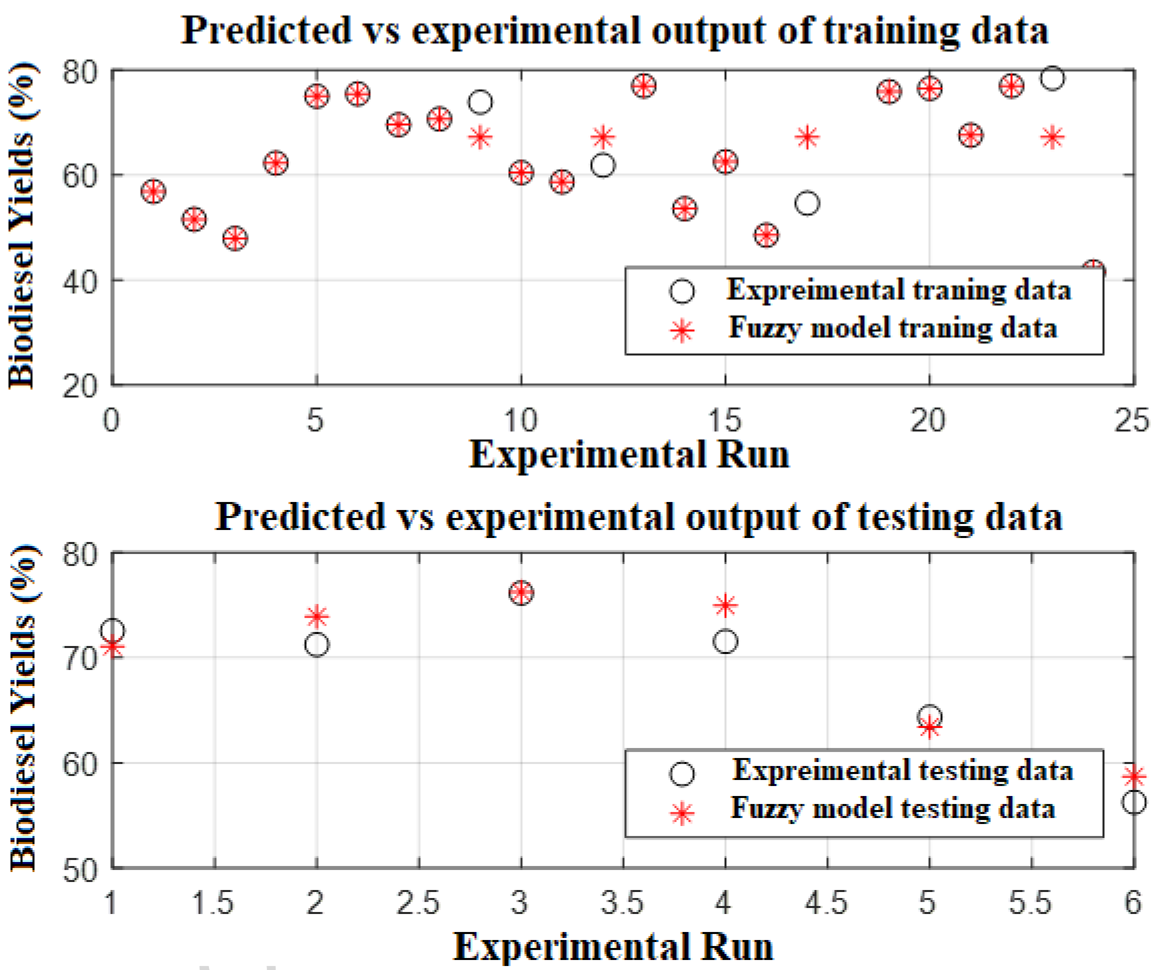

Fig. 2. The plot of fuzzy model predictions against the experimental data

The three-dimensional (3-D) curves for the training and testing phases of the fuzzy model's outputs against the experimental datasets are shown in Figs. 3 and 4, respectively. The figures exhibit the results of every two-input combinations. The results showed good agreement with the experimental data.

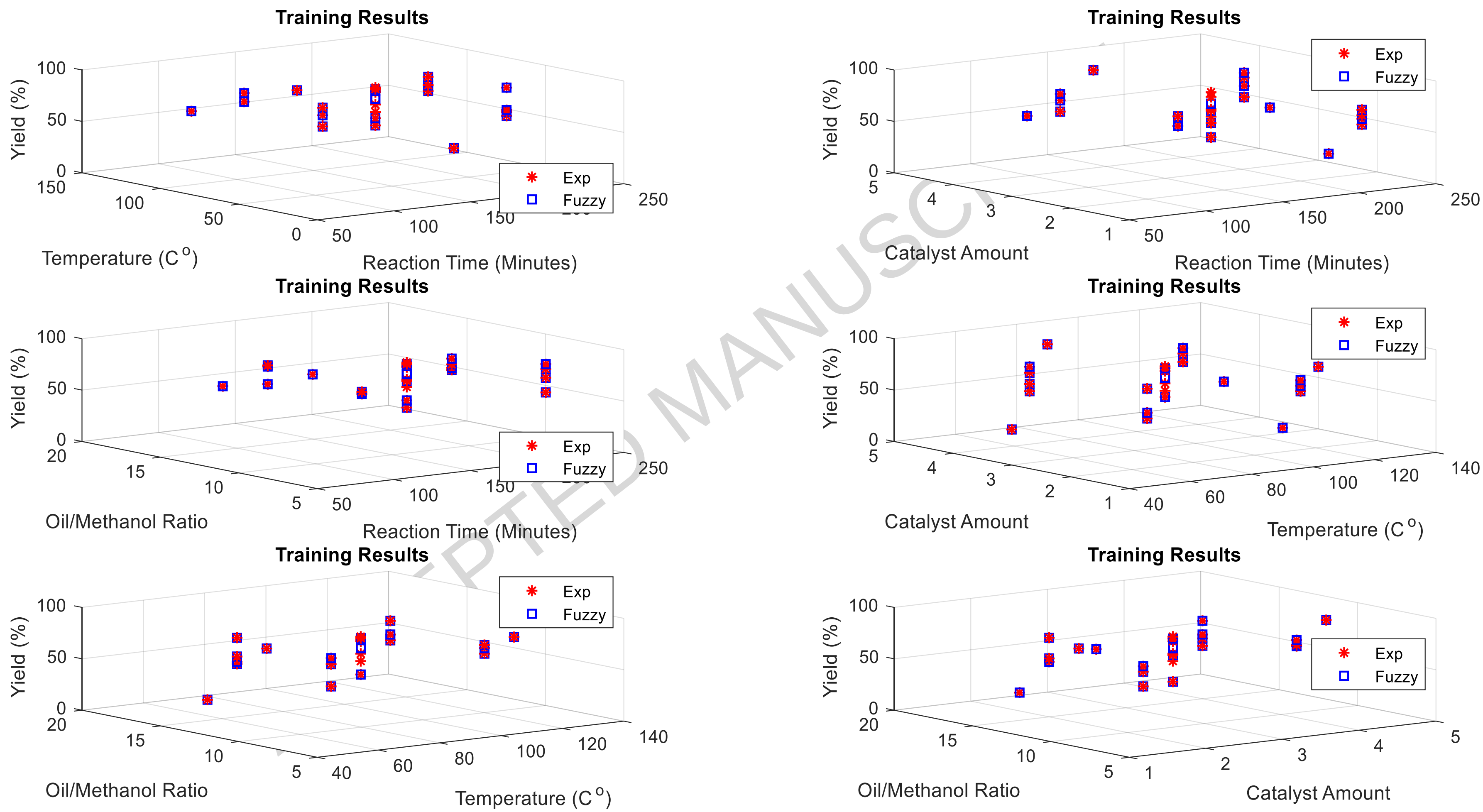

Fig.3. Fuzzy model predictions against experimental data for every two-input combinations of the training phase.

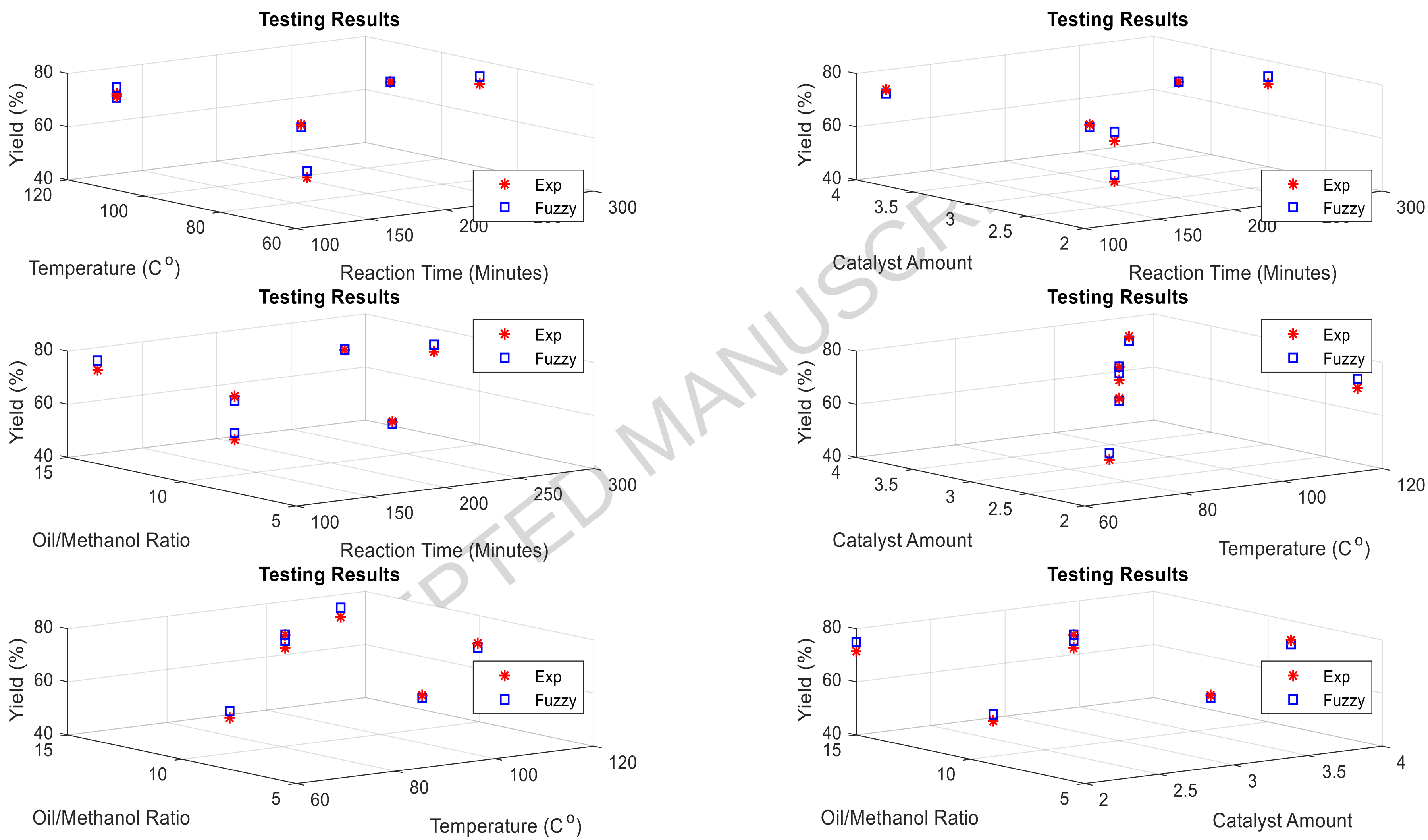

Fig.4. Fuzzy model predictions against experimental data for every two-input combinations of the testing phase.

To illustrate the input-output relationship of the fuzzy model, Fig. 5 presents the 3-D curves of the output in terms of the inputs. However, Fig. 6 shows the MFs of the system inputs. The 3D plot of the reaction time and temperature showed that the increase in temperature (from 40 to 110ºC) is in favor of more biodiesel yield at less reaction time. However, by increasing the reaction time, the biodiesel production is initially increased at low temperature but starts to decrease at high reaction time (more than 150 min). Therefore, a high amount of biodiesel yield can be predicted with low reaction time and high temperature or with high reaction time and a temperature in the range between 65-85ºC. Although more biodiesel yield can be produced with less reaction time and high temperature it will consume more energy as well. The figure shows also the effect of reaction time and catalyst amount where both factors are in favor of biodiesel yield. It appeared that using high reaction time (min) and more catalyst amount (wt%) more biodiesel yield can be produced. It is also observed that a high amount of catalyst produced less biodiesel yield at less reaction time (less than 150 min). On the other side, the 3D results between oil/methanol ratio and reaction time showed that the biodiesel yield decreased with high oil/methanol ratio and high reaction time. This is because using a high amount of methanol will decrease the activity of the catalyst. The results also showed that if the reaction time lasts for more than 200 minutes and the oil/methanol ratio is between 1:12-1:16 this can produce a high amount of biodiesel yield. It is also noted that from the results of temperature and catalyst amount that both parameters proportionally enhanced the biodiesel yield where more than 90% of biodiesel yield can be obtained by increasing them altogether. But from the other side, increasing both parameters also affects the overall cost of the biodiesel production process. This is because to increase the temperature more energy is required and more amount of catalyst is needed which will increase the overall cost of the process. The profile of temperature and oil/methanol ratio shows that at a low value of oil/methanol ratio, there is no prominent change of biodiesel yield occurred when the temperature is increased. However, increasing the temperature for the process at an oil/methanol ratio between 1:14-1:17, the biodiesel yield increased from 57-82%. The 3D trend between oil/methanol ratio and amount of catalyst showed a mixed behavior, as both factors are highly depending on each other. The activity of the catalyst is highly dependent on the amount of methanol in the process for biodiesel production. It is observed that less amount of biodiesel yield is produced at a low amount of catalyst (wt%). Keeping catalyst amount constant at 3 wt%, initially, the biodiesel yield increased by increasing the oil/methanol ratio but after that, it decreased at higher oil/methanol ratio > 1:14.

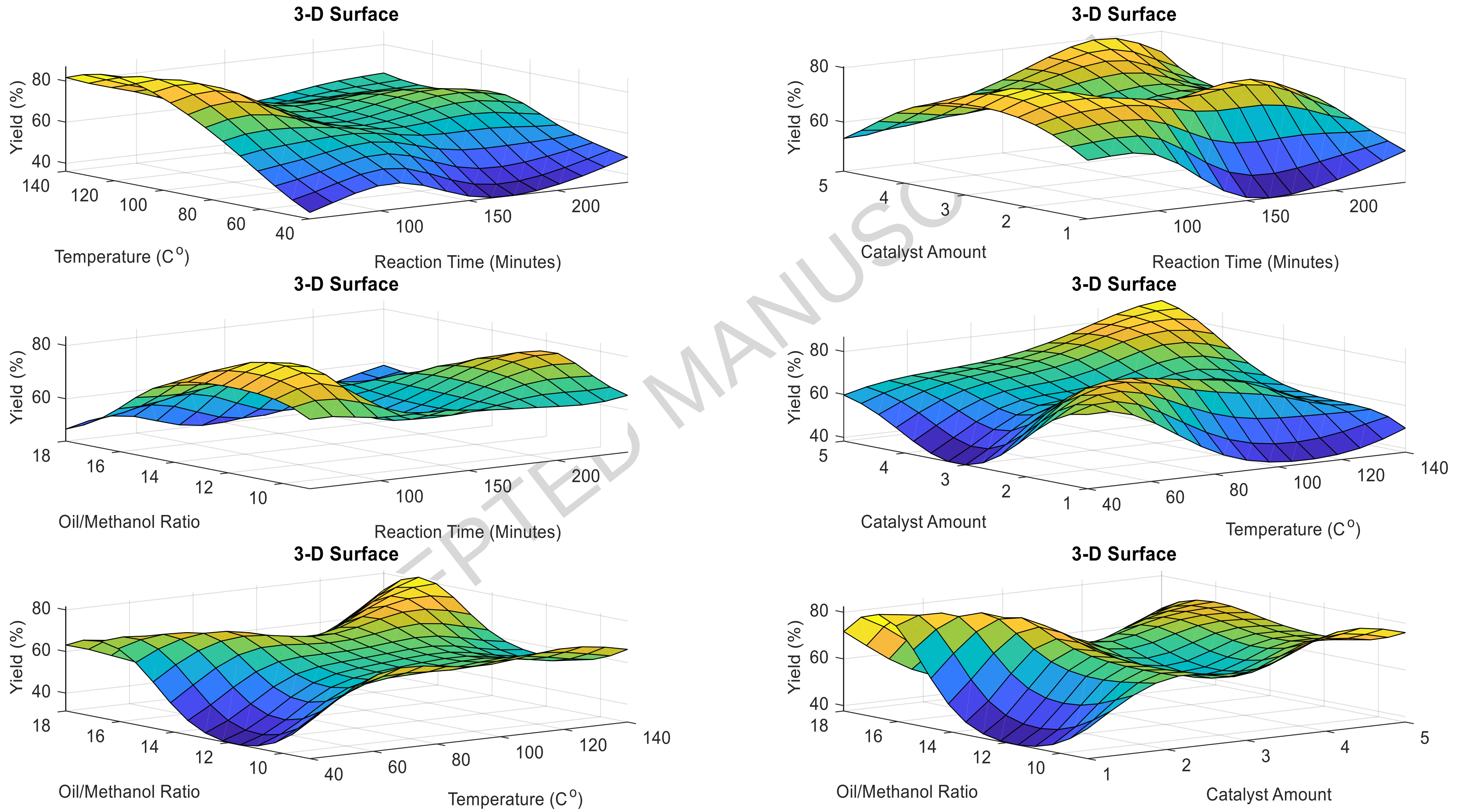

Fig. 5 The input-output relationships of the fuzzy model for every combination of the inputs.

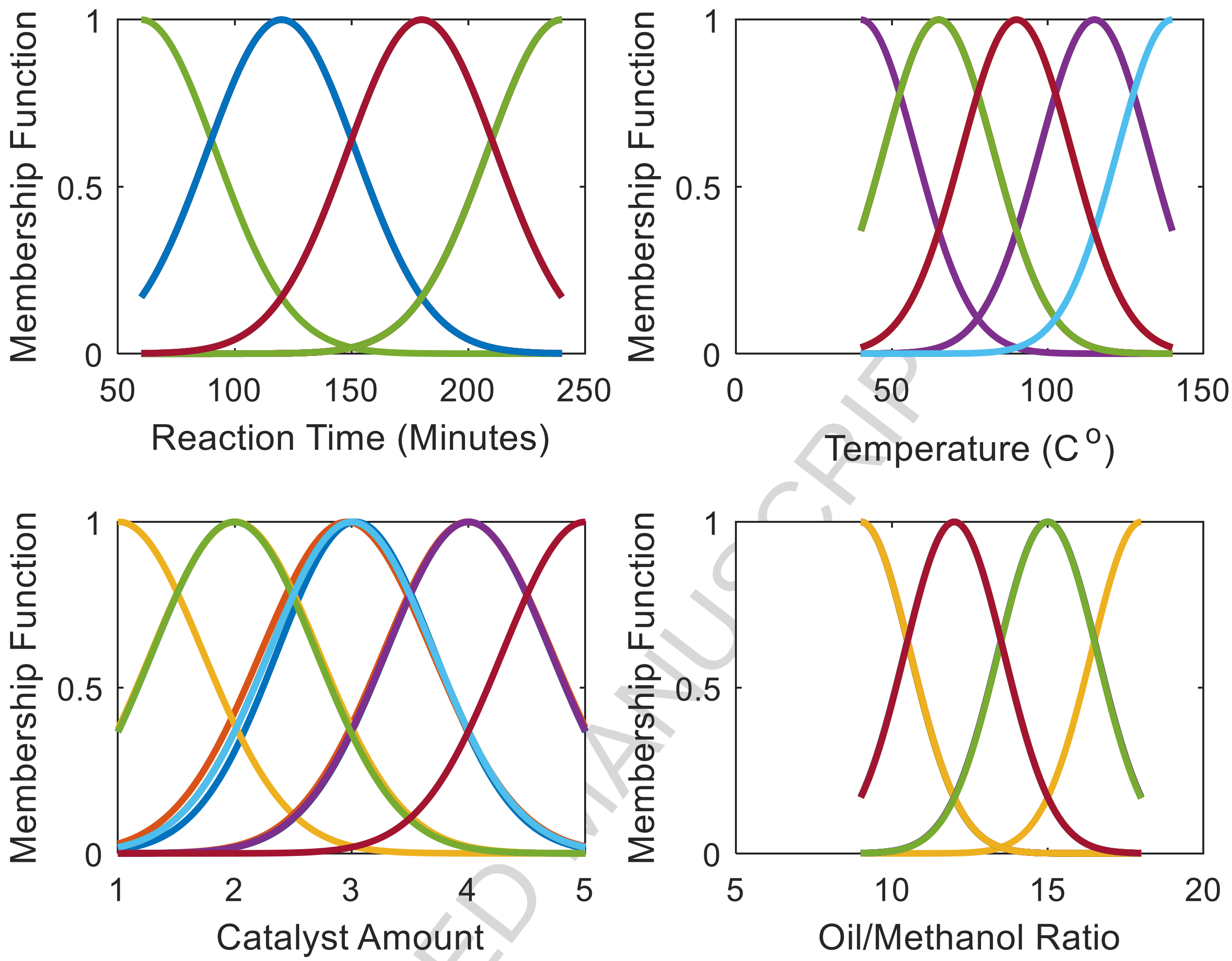

Fig. 6 The biodiesel fuzzy model inputs' MFs

To ensure the prediction accuracy, the predicted output is plotted versus the target for both training and testing phases as shown in Fig. 7. From the figure, it is noticed that the data points of predicted output for both training and testing phases are distributed very close to the line of one hundred percent accuracy. The accurate modeling phase is very mandatory to trust the resulting output of the model for any new or unseen input data.

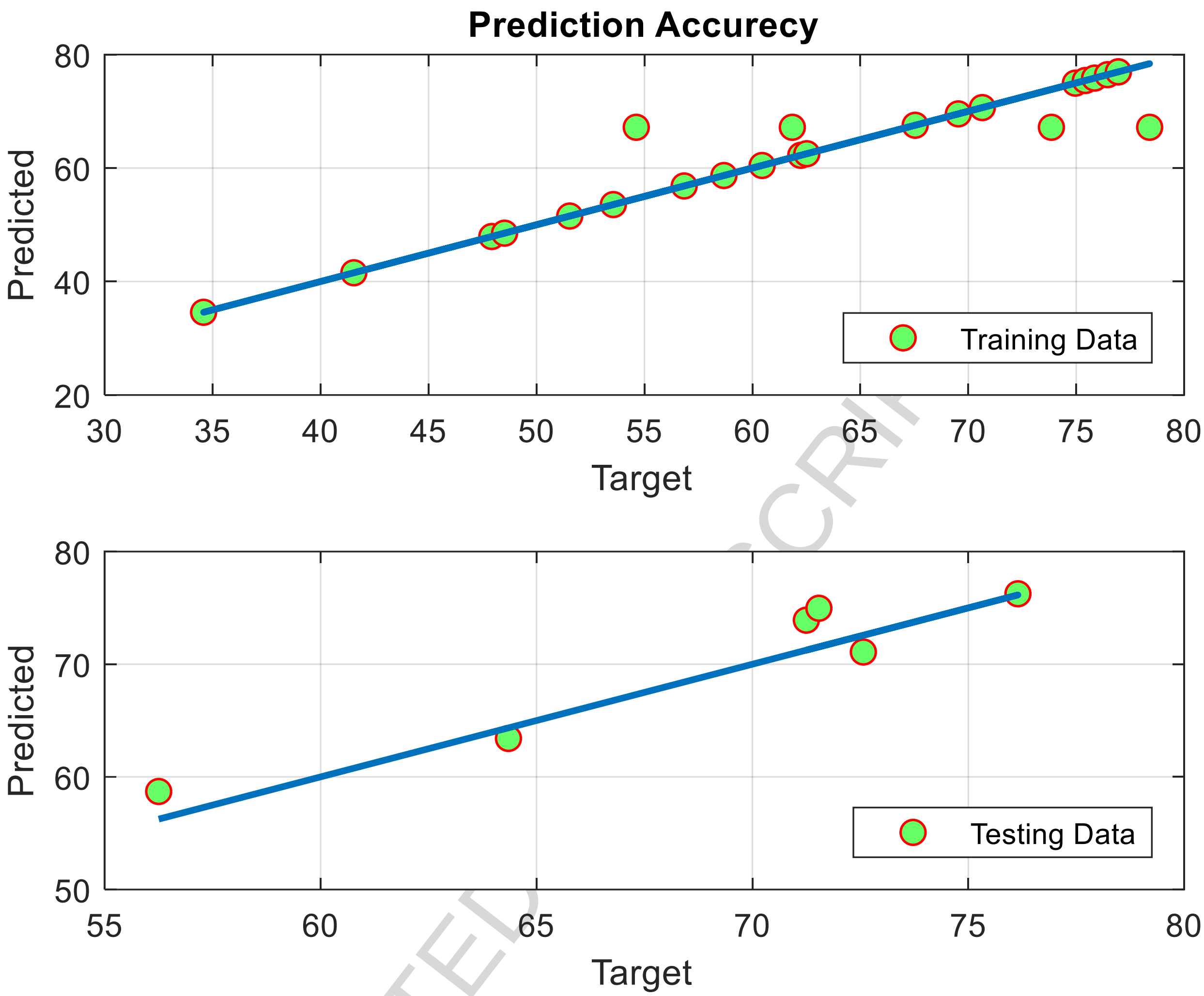

Fig. 7 Prediction Accuracy

The cost function of the optimization process related to the maximum value found so far is shown in Fig. 8.

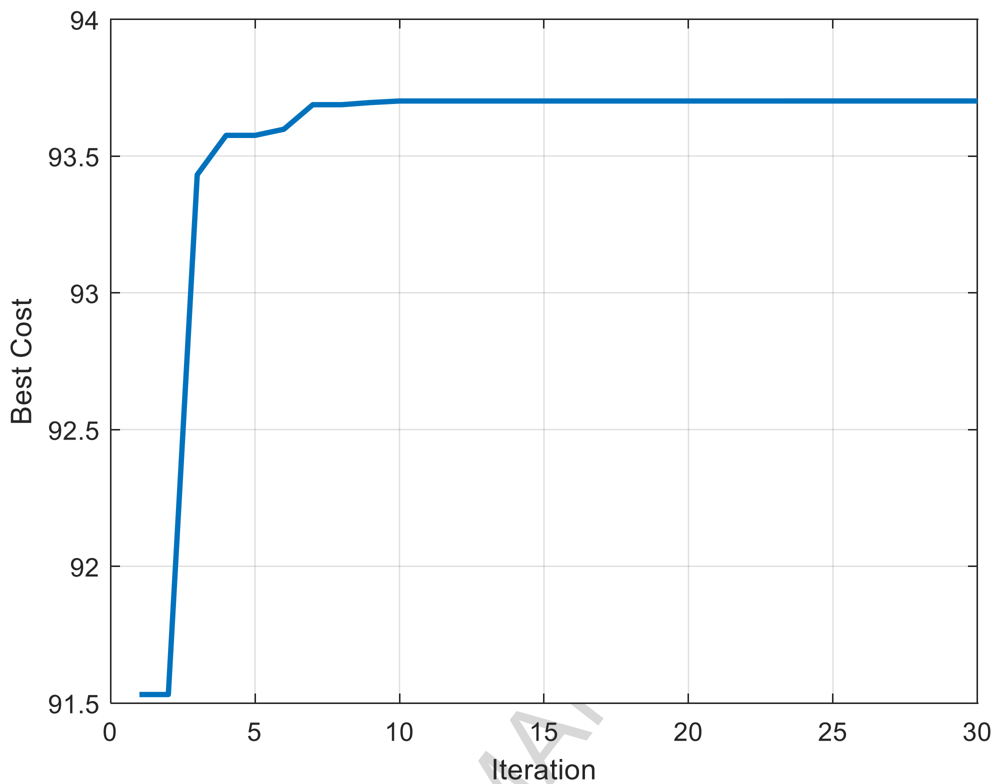

Fig. 8 The optimization curve of the run that had a maximum result of all runs

The resulting optimal parameters and their corresponding predictions are compared with the previous work of Ayoub and his colleagues (2016) as shown in Table 1. As it appeared from Table 1, the percentage of biodiesel production is increased and is better than the obtained in Ayoub et al. (2016) when using PSO as an optimizer. This current study showed that a higher yield of biodiesel can be obtained at a lower temperature (69.66ºC) compared to the earlier work (i.e. 90ºC). Moreover, this work revealed that the more predicted yield of biodiesel from waste frying oil is obtained at a higher amount of catalyst and reaction time while oil/methanol ratio should be less.

Table 1: the resulting optimal parameters compared with the results in Ayoub (2016)

Method	**Reaction Time (minutes)**	**Reaction Temperature (º)**	**Catalyst Amount (wt%)**	**Oil/Methanol (Ratio)**	**Yields (%)**
Optimal as in Ayoub et al. (2016)	180	90	3	1:12	78.40
Optimal using Fuzzy Model-PSO	300	69.66	5	1/9	93.7014

To confirm the results obtained from the optimization process, the optimizer was run for 100 times and the statistical data is shown in Table 2. The statistical data includes minimum (Min), maximum (Max), average (Avg), and standard deviation (StD). In Table 2, the average value is found to be very close to the maximum value and this is supported by the value of the standard deviation StD, which is very small. This will definitely support the results obtained from the optimizer and make it more trustable. The 100 maxima resulting from the optimization process and their average are shown in Fig. 9.

Table 2: The statistical data of the optimal variables and the resulting output after the optimization process

Statistical Operation	Reaction Time (minutes)	Reaction Temperature (ºC)	Catalyst Amount (wt%)	Oil/Methanol (Ratio)	Yields (%)
Min	211.6817,	94.2384,	2.2908,	1/12	83.0657
Max	300	69.6642	5	1/9	93.7014
Avg	209.0517,	89.8205,	4.0615,	1/11	92.1347
StD	94.7078,	29.0722,	1.3519,	1/(3.4543)	2.3878

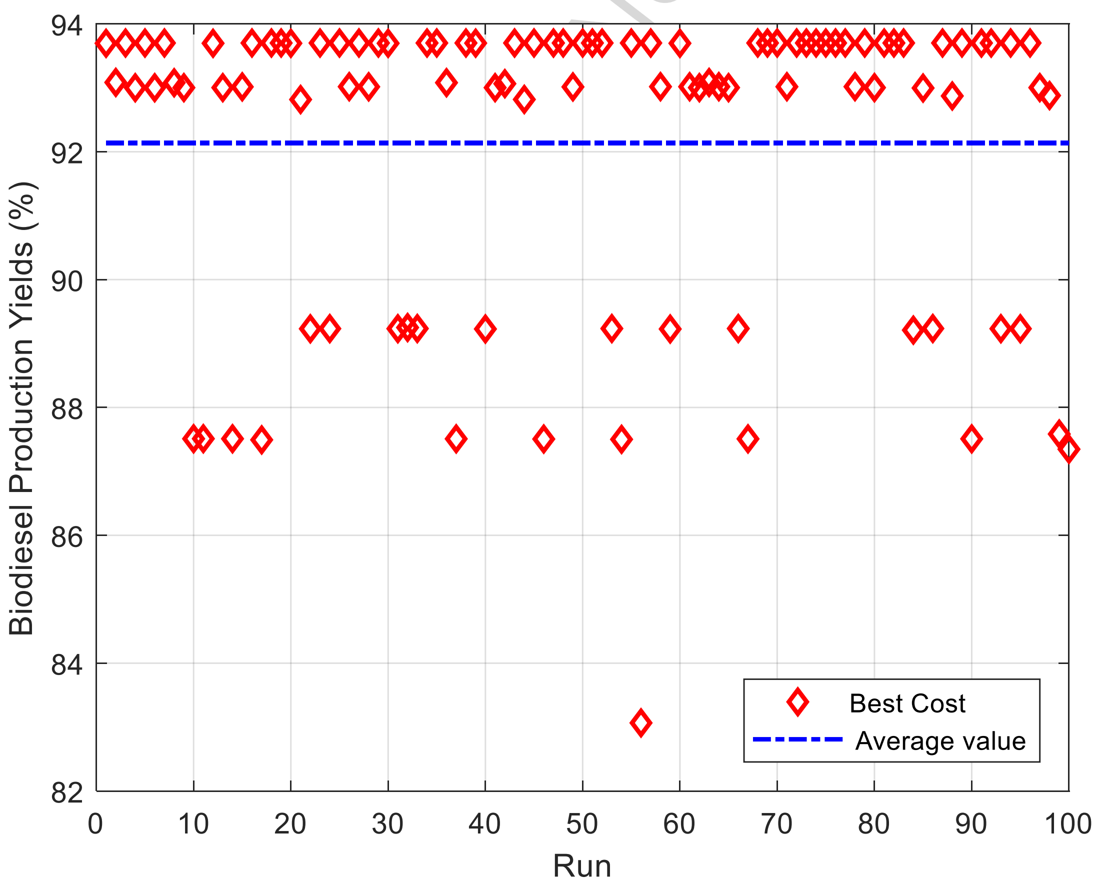

Fig. 9. The cost function values and their average of the 100 optimization process runs

4. Conclusion

A fuzzy logic based model has been presented for producing the biodiesel from waste frying oil via heterogeneous clay based catalyst. Current study enhanced the biodiesel yield by 15% using the transesterification of waste frying oil via swarm optimization (PSO) approach. Four operating parameters of biodiesel production process; temperature, reaction time, oil/methanol ratio and amount of catalyst have been investigated for optimum conditions. Model predictions were validated with the experimental data and showed good agreement. The resulting optimal parameters' values of the biodiesel process using PSO optimizer were a temperature at 69.66ºC, a reaction period of 300 minutes, a methanol/oil ratio of 9:1 and an amount of catalyst of 5 wt%. The obtained results showed that a percentage of 93.70% of biodiesel yield can be achieved at the prescribed optimum operation parameters. The authors can claim that the current developed model could be further scaled up for commercial applications. It is recommended to extend the current research work by using another commercial catalyst at the proposed optimum condition for biodiesel production from waste frying oil.

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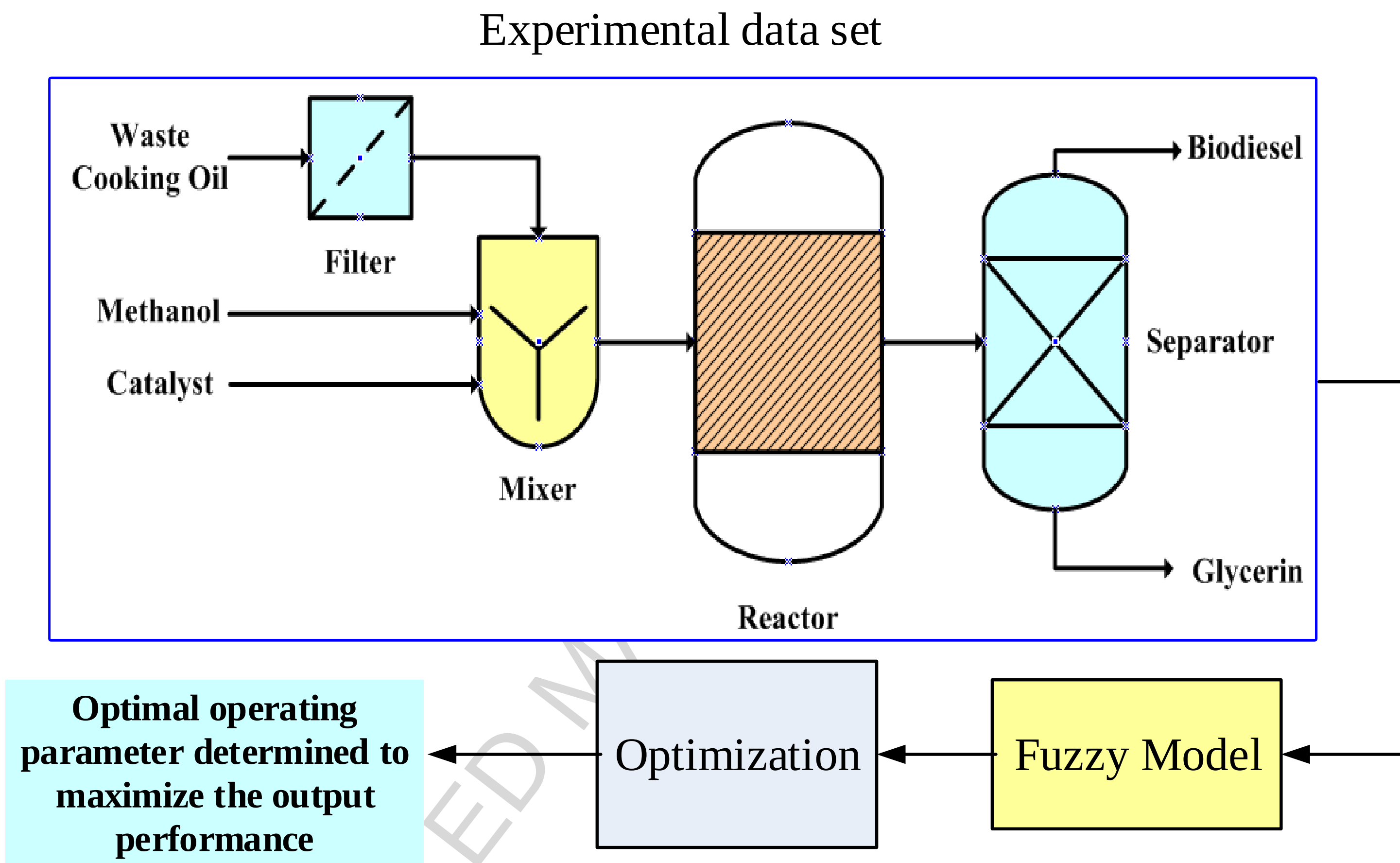
Graphical abstract



Highlights

- Fuzzy logic based model developed for biodiesel production from waste frying oil.
- Particle swarm optimization applied to determine optimal operating parameters.
- Temperature, reaction period, oil/methanol ratio and catalyst amount optimized.
- Biodiesel production enhanced by 15% using proposed optimization strategy.