Optimal operating parameter determination and modeling to enhance methane production from macroalgae

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Abstract- This work aims at proposing a robust strategy to determine the optimal operating parameters based on fuzzy modeling for enhancing the productivity of methane using *Pelvetia canaliculata*. The applied strategy is a combination of fuzzy logic (FL) modeling and particle swarm optimizer (PSO). First, FL is used to build a model that describes methane production using the experimental datasets. Second, a PSO algorithm is used to obtain the best-operating conditions of the production process. The decision variables used in the optimization process are beating time and the feedstock / inoculum ratio (F / I). Each parameter was studied for three different values. The beating time was set at 0, 30, and 60 min while the F/I ratio was set at 0.3, 0.5, and 0.7. To assess the resulting performance, a comparison study was carried out between the optimized results thought proposed strategy and those obtained by using Response Surface Methodology (RSM). The FL model produced a higher accuracy, i.e., lower values of Root Mean Squared Errors (RMSEs), compared with the RSM. Therefore, the obtained results confirmed that the proposed strategy is better than RSM.

Key-words; Renewable energy; Biomethane; Biomass; Algae; Fuzzy logic; Optimization

1. Introduction

Macroalgae is considered as one of the bioenergy sources where it was first investigated in 1973 during US Ocean Food [1]. Macroalgae demonstrated promising biogas productivity [2] [3]. Macroalgae are currently used in food, fertilizer, medicine, and chemical processing industries [4]. Biofuels derived from algae are the so-called "third-generation" and include bioethanol [5] [6], biodiesel [7] [8], and biogas [9]. Some advantages of macroalgae over terrestrial plants are the shorter life cycles, no need for freshwater, and furthermore, no competition for resources with the food industry [10]. While microalgae biofuel production is mainly focused on biodiesel due to the high lipid content of some microalgae species, biogas is the most widely biofuel produced from macroalgae. Integrated production of biodiesel and biogas is being developed using microalgae debris after lipid extraction for anaerobic digestion. Macroalage contains a higher amount of carbohydrates, which can be converted to bioethanol [11]. The major advantage of macroalgae is the mild pre-treatment conditions required compared to second-generation biofuels. Generally, prior to processing the algae for the conversion process to biofuels, lower temperatures, less severe acid conditions, and shorter reaction times are mandatory requirements. Algal biomass is made of organic substance that mainly consists of complex polymeric macromolecules, such as polysaccharides and proteins. Through the anaerobic digestion process, these macromolecules are converted into biogas which is essentially contains methane and carbon dioxide. Generally, the anaerobic digestion process can be summarized as three main consecutive steps: (1) hydrolysis, (2) acetogenesis, and (3) methanogenesis. The hydrolysis is defined as the rate-limiting step in the process, and it depends on several parameters such as the size of the substrate, pH, and the permeability of enzymes to substrate's membranes. In order to increase the surface area available for the enzymatic attack, the algae biomass must be pretreated prior to the anaerobic degradation [12].

Pre-treatment methods for macroalgae biomass are mandatory to obtain better results. These methods can include physical, thermal, chemical, biological, and combined processes. The mechanical method is already used for decreasing the size of the macro-algal, and thus increasing the surface/volume ratio, which results in decreasing the digestion cycle [11]. The methane yield is significantly improved, and reduced conversion times are achieved when filamentous algae *Rhizoclonium* was treated in a warring blender until the particle reaches a size less than 0.1 mm. A further increase was achieved if the mechanically treated samples were further sonicated for 10 minutes [13]. Enzymatic (lipase, α-amylase, xylanase, protease, and cellulase) treatment of *Rhizoclonium* demonstrated high methane productivity. These enzymes can be used in the form of a mixture, or it can be used separately [13]. In the prototype machine owned by TK Energi A/S, the methane potential for *F. vesiculosus* and filamentous red algae are increased after a mechanical pretreatment process is applied. The machine applied pressures up to 1000 bar, which is able to convert a mixture water-algae to a shredded slurry. *Fucus vesiculosus* can produce maximum methane potential when the pre-treatment is followed by the addition of an enzymatic mixture [14]. At the same temperature (11–13% increment), the pre-treatment of red macroalgae *Palmaria palmata* with NaOH at moderate temperatures (20-80°C) is positively influencing the productivity of the methane compared with the standard thermal pre-treatment process [15]. When pre-treated for 10 min on a Hollander beater, *Laminariaceae* spp. attained excess by 52% and 53% in biogas and methane yield, respectively, in a thermophilic range. The same treatment increased the biogas production of *Fucus Vesiculosus Linnaeus* and *Fucus Serratus* from 64 ml/gTS to 181 ml/gTS and from 72 ml/gTS to 230 ml/gTS, respectively [16]. Mechanical pre-treatment has proved to enhance the biofuel yields of other substrates such as waste paper (21% improvement in methane yield) [17], ley silage (59% improvement in methane yield), meadow grass (+24% methane production), switchgrass (improvement in methane kinetics) [18], microalage (18% lipid extraction yield) [19], maize silage and manure [18].

In this study, the effect of two controlling process parameters (inoculum to feedstock ratio and the beating time on a Hollander beater) on the methane yield from *Pelvetia canaliculata* is evaluated. The optimal parameters are identified through PSO based on the model, which has been built using the fuzzy logic technique. First, the experimental dataset has been used to build the model using the FL modeling technique. Second, the PSO algorithm, as one of the simple and fast optimizers, is used for the optimization process.

2. Methodology

2.1. Experimental setup

In March 2016, the macroalgae *Pelvetia canaliculata* were collected on-shore in Rothesay (Scotland). The inoculum was provided by Energen Biogas Plant (Cumbernauld, Scotland), the plant used food residues as feedstock. Both algae and inoculum were stored at 4°C and used within 48 h [20]. The general characterization of the sludge and algae used in this study is shown in Table

1. The algae were pre-treated in a Hollander beater under the effect of the shear stress between the rotating bladed drum and the bedplate. The exerted pressure is controlled through the adjustment of the distance between the blades and the bottom plate. The beater had 40 kilograms of water and 0.9 kilograms of algae. The sample's pre-treatment was processed at 30 and 60 min.

Table 1. Inoculum and macroalgae characterization

Parameters	Inoculum	Macroalgae
Total Solids (%)	4.70 ± 0.01	18.7 ± 0.01
Volatile Solids (%)	62.98 ± 0.09	81.68 ± 0.06
Ash content (%)	37.02 ± 0.09	18.32 ± 0.06
C (% of TS)	-	37.09 ± 0.01
H (% of TS)	-	5.41 ± 0.01
N (% of TS)	-	2.48 ± 0.01
O (% of TS)	-	37.51 ± 0.01

Biochemical methane potential (BMP) tests were carried out as specified in [11] according to standard procedures [21]. The reactors were supplied with a constant amount of inoculum, and the quantity of pre-treated algae pulp was adjusted to achieve the required F/I ratios (0.3, 0.5, and 0.7). The pH was adjusted to 6.70±0.15 with potassium dihydrogen phosphate (KDP) as a buffer solution. The digestion process was terminated as long as the daily biogas production rate was decreasing. In other words, the rate is decreasing to reach a value of less than 1% of the overall obtained volume [21]. A temperature of 0 °C and a pressure of 1 atm are considered as the standard conditions to give the methane volumes for dry gas.

2.2. Fuzzy logic (FL)

Artificial intelligent (AI) technology has a great impact on the systems' modeling due to two reasons. First, it is considered as a general approximator for linear and nonlinear systems. In other words, it can efficiently model a signal that has a high complexity between the output and its corresponding inputs. Second, it has the ability to learn from the input-output data samples and hence updates the system's parameters accordingly in order to improve its performance.

AI techniques are proved to be efficient modeling tools as they have the ability of learning. FL modeling is considered as one of the most efficient AI modeling tools where it is capable of tracking the trends of data precisely with a small number of training epochs [22]. Fuzzification, inference system, and defuzzification are the main stages of the FL modeling. In the fuzzification, stage, the inputs are transformed from their crisp values to the corresponding fuzzy values via a mapping function, namely membership function (MF). The most popular are the Gaussian and the triangular shape functions. As soon as the inputs have been fuzzified, they are fed to the inference system in the second stage to fire the fuzzy rules. Usually, the rules are built either by an expert or from the input-output data. The first method is popular in fuzzy control systems; however, the second is used in fuzzy modeling as in the current case. There are many algorithms to extract the fuzzy rules from the input-output data. The most famous one is the "Subtractive Clustering" method, adopted in the present research. This method partitions the input-output space into different clusters, and by using an optimization algorithm, it relates the inputs-space to the output-space in the form of an IF-THEN rule.

In fuzzy logic, there are two well-known fuzzy rule forms. The Mamdani-type and the Takagi-Sugeno-Kang-type. Sometimes, the latter is abbreviated to TSK-type or Sugeno-type. The fuzzy rule of a two-input one-output system takes the form as in Equations (1) and (2) for Mamdani-type and Sugeno-type, respectively:

$$\text{IF Input}_1 \text{ is in } \mathrm{MF}_{in1} \text{ and Input}_2 \text{ is in } \mathrm{MF}_{in2} \text{ THEN Output is } \mathrm{MF}_{o} \quad (1)$$

$$\text{IF Input}_1 \text{ is in } \mathrm{MF}_{in1} \text{ and Input}_2 \text{ is in } \mathrm{MF}_{in2} \text{ THEN Output} = f(\text{Input}_1, \text{Input}_2) \quad (2)$$

where MF_{in1} and MF_{in2} are the membership functions of input 1 and input 2, respectively, and $f(.)$ is a function of the inputs which could be linear or nonlinear.

The outputs of the rules are aggregated together to produce the final fuzzy output. Then, this output is defuzzified to its corresponding crisp value. The Centre of Gravity (COG) and Weighted Average are the two famous defuzzification methods in the case of Mamdani-type and Sugeno-type, respectively. More details about the best fuzzification and defuzzification methods as well as the FL modeling, can be found in [23-26].

The methane yield from macroalgae could be enhanced through optimizing the beating time and feedstock/inoculum ratio. The methane yield production under-considered optimal controlling parameters has been studied in our previous work [20]. With different operation conditions, twelve different experiments were conducted. The Implementing Design of Experiment methodologies, modeling, and optimization were carried out using the set of the collected data.

In a real experiment, the collected data is highly expected to be superimposed with noisy signals. This results in a dataset that contains uncertain data values. In this case, the fuzzy logic technique is the best to produce a robust model. In our previous study using ANOVA [20], the obtained model is built using a highly nonlinear data set. To obtain a better model, in this work, we have used the fuzzy logic tool to build the model. The model is constructed using a two-input and single-output set of the 12 experiments which were conducted in our previous study [20] where an optimum methane yield of 283 ml/gVS was obtained for 50 min pretreatment time and a ratio F/I of 0.3, which represents an increase of 45% compared to non-pretreated algae.

Before starting the training phase, the training data samples were randomly selected from the whole set, and the remaining samples were reserved for the testing phase. The training set has 8 data samples, while the remaining 4 samples were assigned as the testing and validation stages. Figure 1 shows the FL model structure of the methane yield process. In the figure, "Sug21" refers to a fuzzy system of Sugeno-type with two inputs and one output.

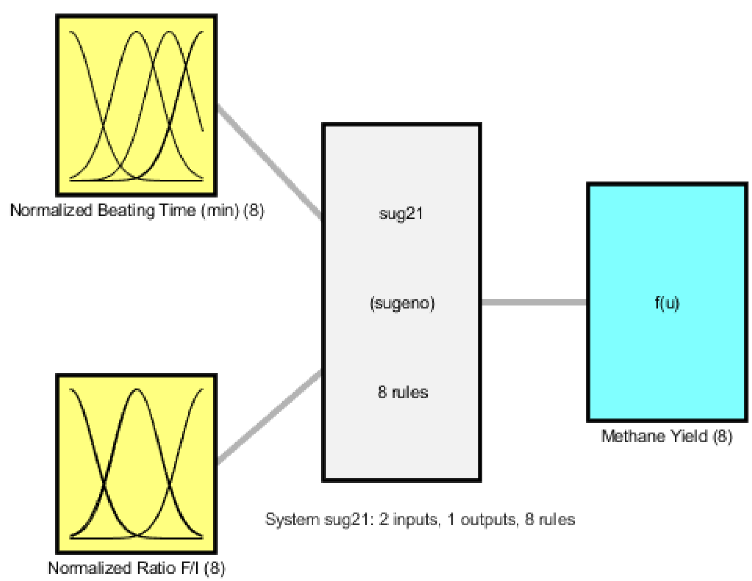

Figure 1. Structure of the FL model

2.3. Particle swarm optimization (PSO)

Some living creatures inspired researchers to emulate their ways of movement. This movement is usually performed in a swarm. The algorithms that describe these procedures are called swarm optimizers. One of these optimizers is the PSO, which mimics the movement behavior of a swarm of birds. This algorithm, like many other optimization algorithms, starts by suggesting some solutions, typically called particles [27]. During the optimization process, the particles are modifying their orientations and locations in an iterative process. The next movement of the particle is calculated based on its best position as well as the best position found so far of the whole swarm's particles [28]. For every particle in the swarm, the next velocity vector Vel^{k+1} and the next position vector Pos^{k+1} are calculated based on the previous velocity Vel^{k} and the previous position Pos^{k}. Equations (3) and (4) are describing the updating rules of the position and velocity vectors, respectively.

$$Pos^{k+1} = Pos^{k} + Vel^{k+1} \quad [29] \quad (3)$$

$$Vel^{k+1} = w * Vel^{k} + c_1 * r_1 * (Pos^{LocalBest} - Pos^{k}) + c_2 * r_2 * (Pos^{GlobalBest} - Pos^{k}) \quad [30] \quad (4)$$

where, w denotes the weight of inertia; c_1 and c_2 denote the self experience weight and the social experience weight, respectively; r_1 and r_2 are two random generators changing from 0 to 1 .

Beating time and FI ratio are two independent controlling parameters that are controlling the methane yield. Therefore, they are selected as the decision variables in the optimization process, and methane yield is the cost function required to maximize.

The flexibility to form hybrid tool optimization tools, the easy implementation , the few parameters to adjust required and the use of simply logic and mathematical operations are the main advantages of PSO. Furthermore the ability to handle functions with probabilistic nature and the ability to start the iteration process even with a bad initial solution makes PSO a powerful technique for the optimization of this type of processes.

3. Results and discussion

Two input parameters were studied at three levels: the beating time was set at 0, 30, and 60 min, while the feedstock/inoculum ratio was fixed at 0.3, 0.5, and 0.7. The response variable was the methane production in terms of ml per g of volatile solids (ml/gVS). Biochemical methane potential test results from the Response Surface Methodology are shown in

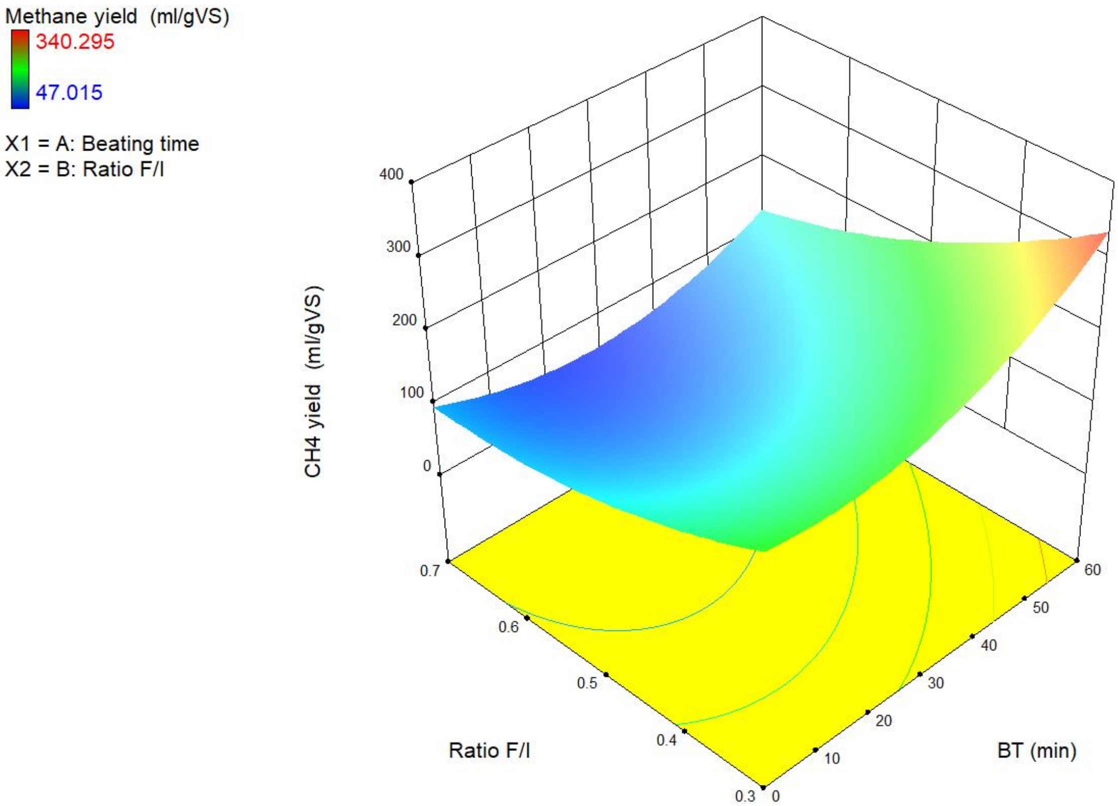

Figure 2. Response surface plot for methane production from RSM.

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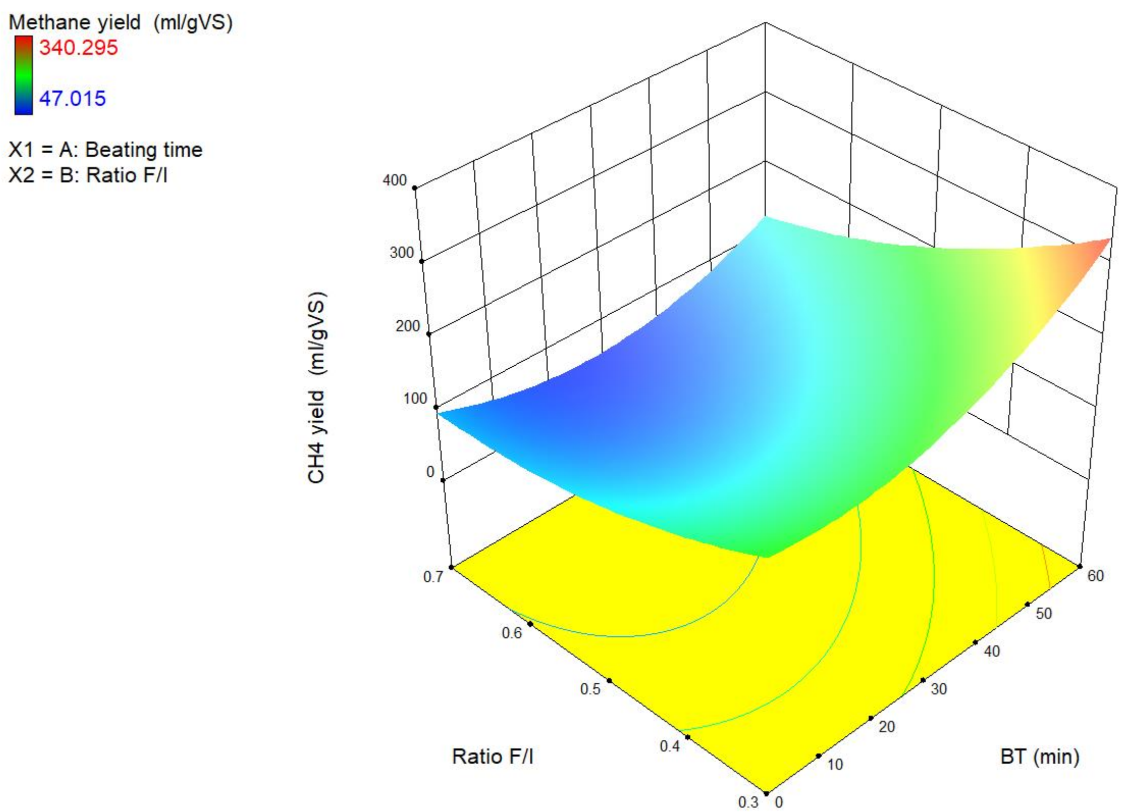

Figure 2. Response surface plot for methane production from RSM.

The fuzzy model had 8 rules which take the IF-THEN Sugeno-type form, as mentioned before. Each rule describes a unique relationship between the system's output and inputs within the input-output space. The following is the form of the m^{th} rule used in this work:

Rule #m: IF "*Beating-Time*" is in "*Input 1 Cluster #m*" and "*Ratio F/I*" is in "*Input 2 Cluster #m*" THEN "*Methane Yield*" is in "*Output Cluster #m*"; where, $m = 1, 2, \ldots, 8$

The system is usually trained by passing a training batch (input-output samples) to the model and then calculating the output's error. Based on this error and using a training algorithm, the model's parameters are changed accordingly in order to minimize the system's error. In the AI field, the training batches are referred to as the training epochs. In the current case study, the number of epochs is set to a value of two.

In fuzzy systems, the membership function (MF) is a function used to map the values of the input variables to their corresponding fuzzy values. This mapping function is the initial step of the fuzzification process. The Gaussian and triangular shapes are the most popular MFs. The former is adopted in this study as shown in Figure 3. In the figure, the word “normalized” referred to the value of the scaled value of the input. To unify the weights of the inputs during the training phase, the values of the inputs are scaled (normalized) to be in the range [0 1].

The adequacy of the RSM model was tested through ANOVA, the statistical significance of the model’s terms and the model itself is examined using the lack- of-fit test and the sequential F-test. The model is considered adequate if Prob. > F for the model and each model’s term does not exceed the level of significance ($\alpha = 0.05$ in this case).

During the FL modeling, the training process is continued until reaching satisfying test results of RMSE and MSE. The RMSE value is found to be 3.8122 for the fuzzy model; however, it was 8.1294 for the ANOVA model [20]. Accordingly, the RMSE is decreased by 53% using the fuzzy model compared to the ANOVA. Additionally, the value of the coefficient of determination for the fuzzy model is found 0.99775, while it was 0.99074 for the ANOVA. From the resulting values of the statistical markers, the fuzzy model is proved superior, which produced less RMSE and high R^2 values over their correspondings of the ANOVA model.

Table shows an accuracy comparison between the results of fuzzy and ANOVA, while the validation results are shown in Table 3. As seen from the two tables that the accuracy of the results obtained from the FL model is better than those of the ANOVA.

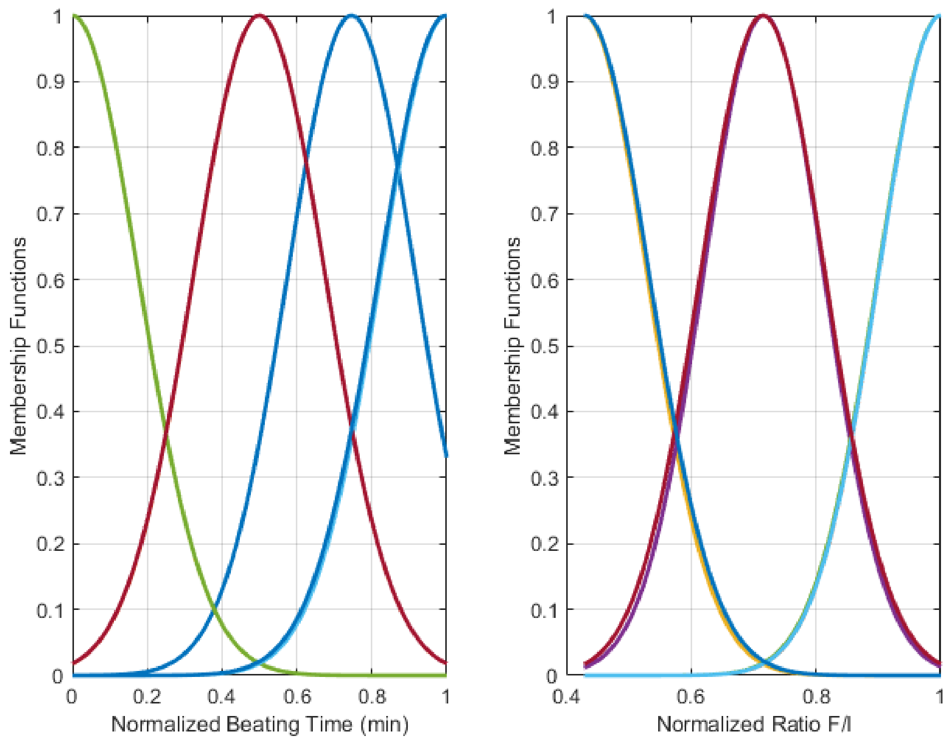

Figure 3. Normalized inputs' MFs of fuzzy model

Table 2. Accuracy of the FL model in comparison with that of the ANOVA

Model type	MSE	RMSE	R^2	Validation Data MSE
Fuzzy	14.5326	3.8122	0.99775	9.2989
ANOVA [20]	66.0872	8.1294	0.99074	31.9337

Table 3. **V**alidation results of FL model in comparison to that of the ANOVA.

Exp.	Inputs		Actual Methane yield (ml/gVS)	Predicted ANOVA [20]	Predicted Fuzzy	% Error ANOVA [20]	% Error Fuzzy
	Beating time (min)	Ratio F/I					
1	20	0.4	130.38	137.53	126.07	-5.484	3.307
2	35	0.6	80.73	77.16	80.66	4.422	0.085

Figure 4a illustrates the resulting predictions obtained using the fuzzy model in this study in comparison with the experimental data and the optimized results using the ANOVA. The experimental runs resulted from setting the beating time at 0, 30, and 60 min and the F/I ratio at 0.3, 0.5, and 0.7, the experimental table is specified in [20]. By deep investigations of the plots, it can be seen that the predictions from the built model using fuzzy logic are almost coinciding with the experimental data to a large extent, which indicates that the fuzzy model is reliable relative to the case of the ANOVA model.

Methane yield for 60 min mechanical pre-treated algae increased by 74% compared with untreated algae, while the increment for 30 min beaten samples was 6%. The pretreatment clearly starts to be effective at beating times higher than 30 min, increasing the surface area of the biomass, which makes it readily accessible to the microorganisms. It is found that the hydrolysis of the feedstock can be accelerated by an excessive particle size reduction of the substrate, which can result in the accumulation of volatile fatty acids (VFAs), leading to the process inhibition and stopping the methane production [9, 18, 31]. This seems not to happen in this study even at the highest pre-treatment times, as the methane yields are maximum at the highest beating time regardless of the feedstock/inoculum ratio. Even that the particle size was not measured against the beating time, it can be concluded that pre-treating the macroalgae *P. canaliculata* for 60 min in a Hollander beater does not reduce the algae particle size to such extent to lead to a digestion inhibition. VFA accumulation due to excessive particle size reduction was not found in previous studies of different macroalgae species [11, 32]. Results from Tedesco et al. showed a maximum biogas yield for 10 min pretreatment lower than the obtained in the present study, confirming that higher beating times have a positive effect on the methane production [32].

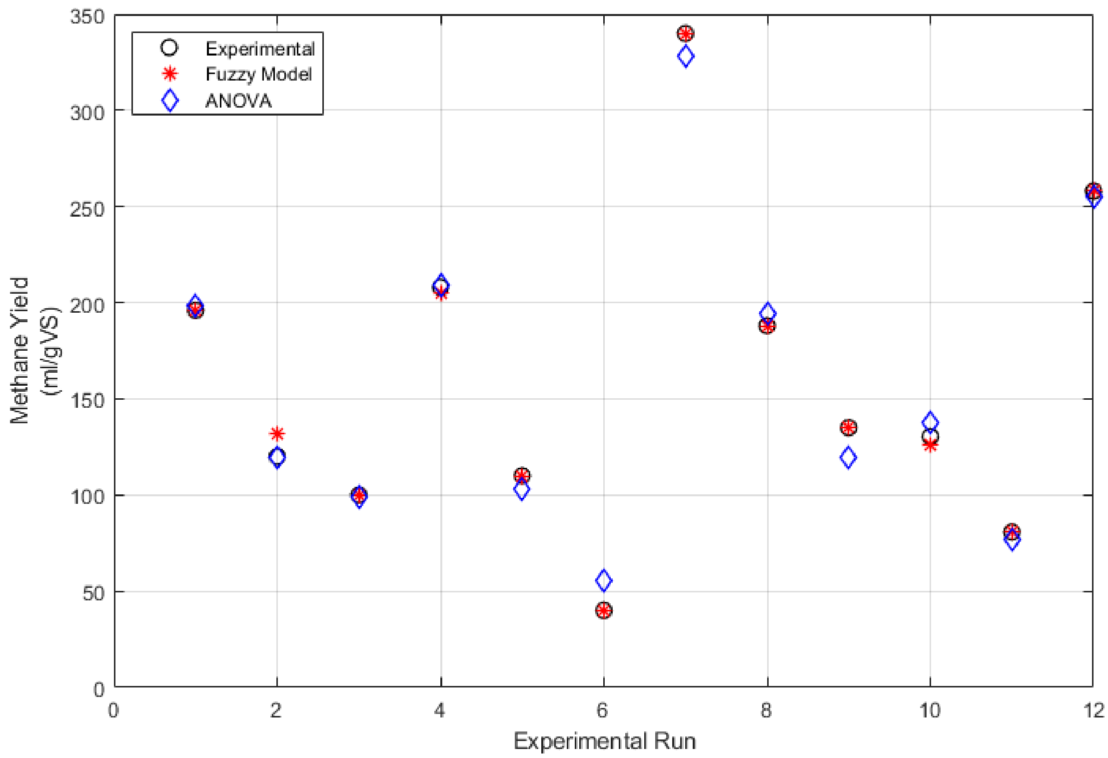

Figure 4a. FL output against the experimental data and ANOVA

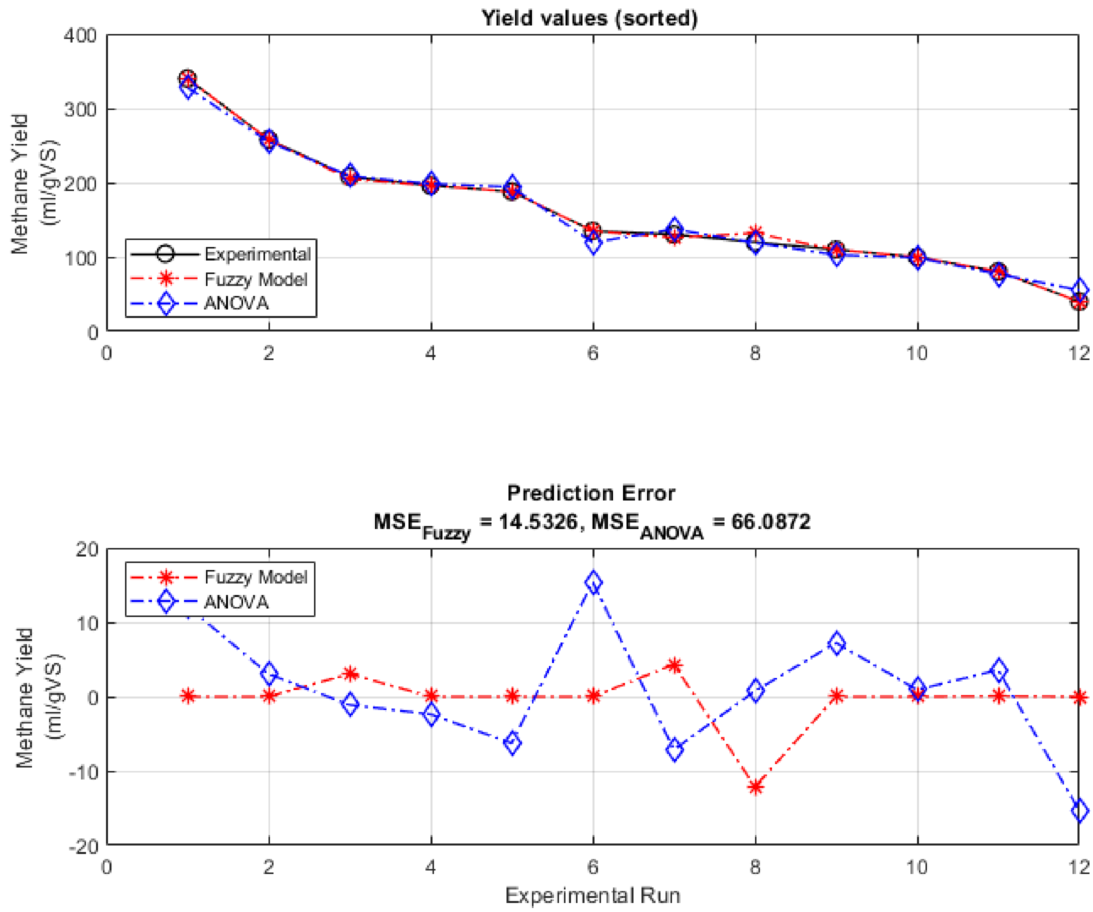

Figure 4b. Prediction error for ANOVA and FL model.

The methane yield versus the normalized inputs data in 3-D spatial shape is shown in Figure 5. As seen from the figure, the methane yield has a nonlinear relationship with the contributions. Both process parameters have an exponential effect on the methane yield. The methane yield is exponentially increasing with the increase of the pre-treatment time, while the effect of the F/I ratio is the opposite; higher F/I ratios resulted in lower methane yields. The effect of pre-treatment time in the methane production is more accused at lower F/I ratios, as shown by comparing the slopes of the 3D plot at low and high F/I ratios. Methane yield is increasing with the decrease of the F/I ratios, both for untreated and pre-treated samples at 30 and 60 min. Non treated algae at a F/I ratio of 0.3 resulted in a methane yield of 196 ml/gVS, while for a F/I ratio of 0.7, the methane production was 100 ml/gVS. According to literature, an optimum F/I ratio for anaerobic digestion is around 0.5 [16], the present study shows that the F/I ratio can be further decreased to 0.3 for the digestion of *P. canaliculata* macroalgae. Keeping the industrial anaerobic reactors to operate at the optimum F/I ratio permits a better manipulation of the biomass and prevents the undigested material leaving the reactor as digestate. Similar results were achieved for the anaerobic digestion of pig urine and rice straw where the lowest F/I ratio resulted in a better operation performance both in terms of biogas production and volatile solids reduction [33]. Usually, high F/I ratios can cause an accumulation of VFAs, resulting in the process inhibition (the same effect produced by excessive communication) [34]. In the case of *P. canaliculata*, even at the highest F/I ratio of 0.7 studied, no inhibition is produced.

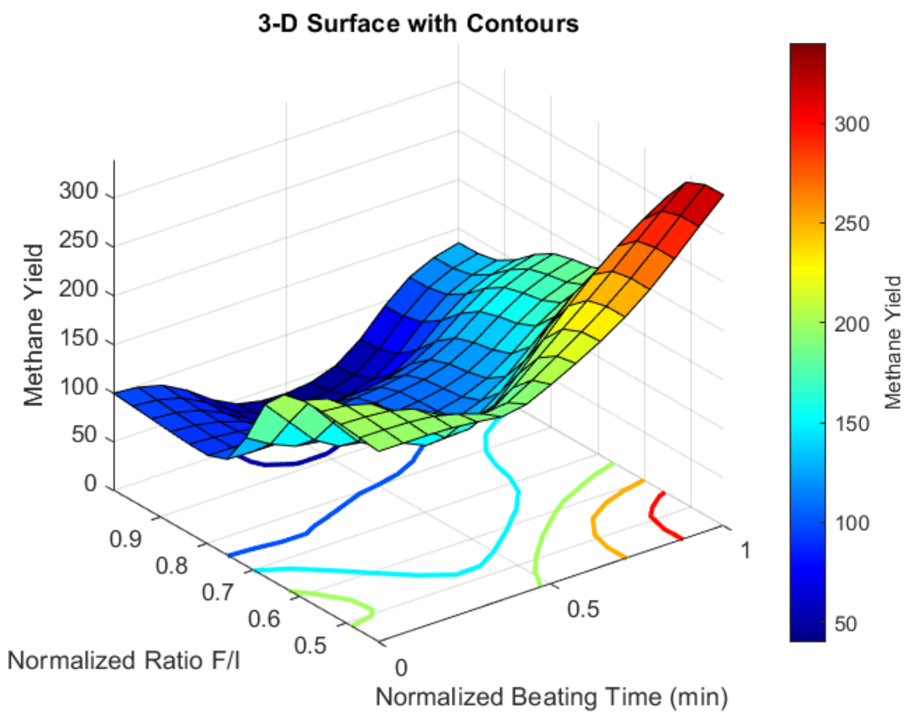

Figure 5. The methane yield versus the normalized inputs data in 3-D from PSO.

Comparing the surfaces plots from both techniques, PSO's plot (Figure 5) showed a higher accurary fitting the experimental data compared to RSM's plot (Figure 2). Even the shape of the surface is similar in both techniques, PSO is able to identify depressions (e.g at high F/I ratio and medium BT) and peaks (e.g. at low both F/I ration and BT) that RSM showed as smooth surface.

In the modeling field and to trust the resulting output of the model, testing the modeling accuracy is a necessity. This can be done by feeding the model with new or unseen data and then investigating the prediction accuracy. To measure the prediction accuracy, the predicted output is plotted versus the target, as illustrated in Figure 66. It can be noticed from the plots that the data of the resulting fuzzy model output is distributed very close to the diagonal line that represents the one hundred percent accuracy. The very high correlation between predicted and experimental values demonstrated the suitability of the fuzzy methodology to model the methane production from mechanically pre-treated macroalgae. This proves the high accuracy of the FL model in tracking the data.

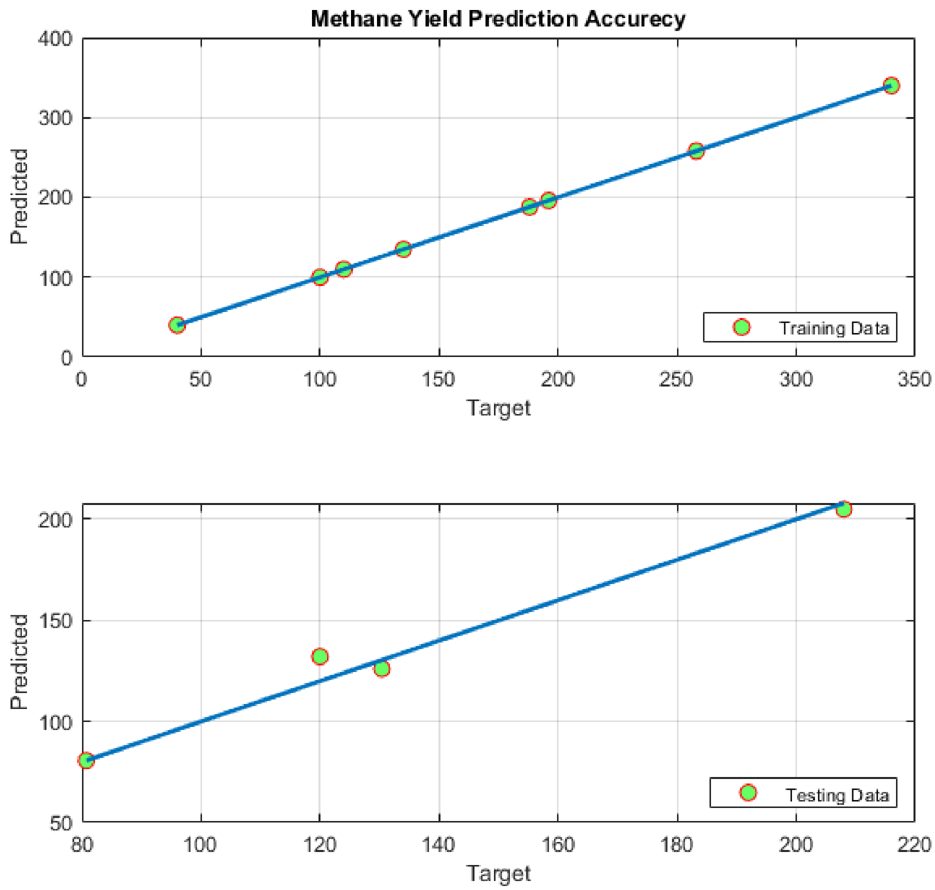

Figure 6. Prediction Accuracy

The plot of the cost function, related to the best (maximum) value of the optimization processes found so far through the one-hundred runs, is illustrated in Figure 77.

The movements of the ten solution particles are recorded during the thirty iterations optimization process to study the particles' convergence. Figure 8a and 8b present the convergence curves for the solutions with the optimizing variables of the beating time and ratio F/I, respectively.

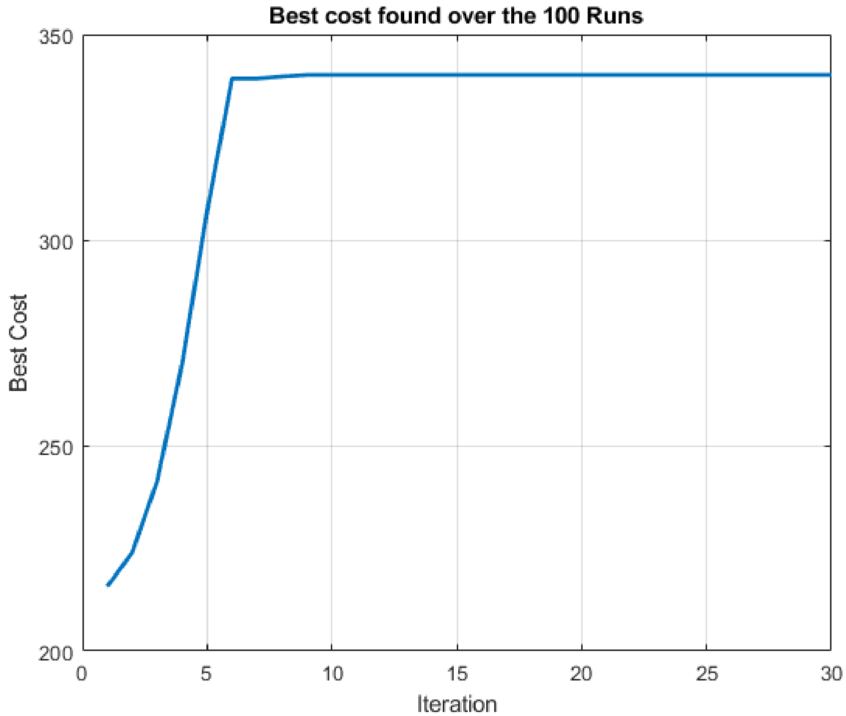

Figure 7. Cost function variation versus iteration during the optimization process.

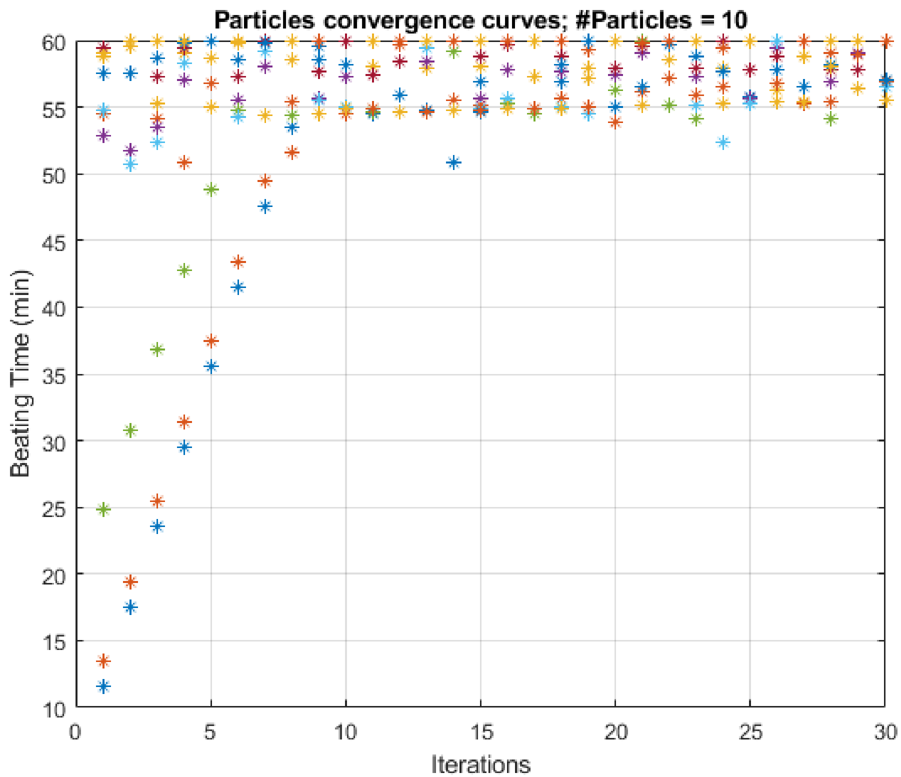

Figure 8a. Particles' convergence plots for the beating time.

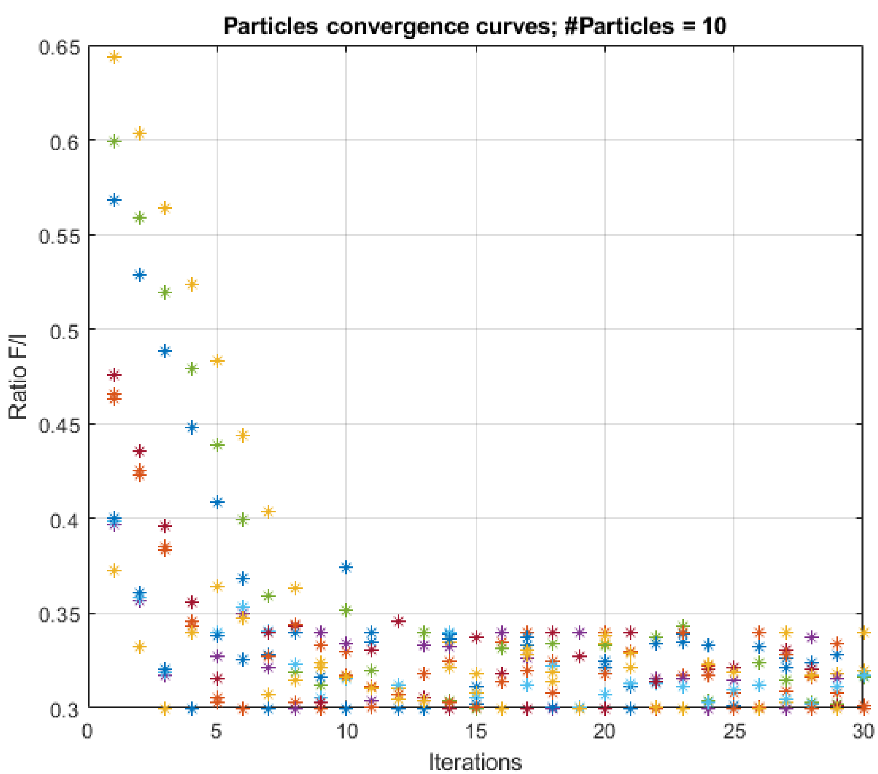

Figure 8b. Particles' convergence plots for the ratio F/I.

One of the features of the Fuzzy model is the ability to predict values outside the training range. Accordingly, the optimization process search space of the inputs is expanded by certain different percent below and above the lower and upper bounds, respectively. The obtained optimization results are presented in Table 4. It can be noted from table 4 that the extension beyond the training range of the inputs improves the methane yields, especially that of the beating time.

Table 4. Optimized results

The extension (%)	Statistical Measure	Methane Yields (ml/gVS)	Associated Optimal Inputs	
			Beating time (min)	Ratio F/I
0	Min	204.1299	0	0.4038
	Max	340.1674	60	0.3096
	Avg	323.8427	52.8	0.3210
	StD	44.4294	19.5959	0.0310
	RMSE	1.2761e+02	5.6285e+01	8.8394e-02
5	Min	204.1301	0	0.4039
	Max	355.4615	63	0.3064
	Avg	340.3283	56.7	0.3163
	StD	45.6280	18.9952	0.0295
	RMSE	1.4357e+02	5.9767e+01	9.2426e-02
10	Min	204.1302	0	0.4040
	Max	369.3012	66	0.3040
	Avg	362.6942	63.36	0.3081
	StD	32.5298	12.9985	0.0197
	RMSE	1.6183e+02	6.4667e+01	9.7877e-02
15	Min	204.1301	0	0.4039
	Max	381.7298	69	0.3022
	Avg	369.2976	64.17	0.3093
	StD	45.5421	17.6938	0.0262
	RMSE	1.7127e+02	6.6541e+01	9.8159e-02
20	Min	204.1299	0	0.4038
	Max	392.9587	72	0.3009
	Avg	377.8523	66.24	0.3091
	StD	51.4859	19.6315	0.0282
	RMSE	1.8112e+02	6.9060e+01	9.8771e-02
25	Min	204.1288	0	0.4053
	Max	403.2548	75	0.2999
	Avg	399.2720	73.5	0.3020
	StD	28.0180	10.5529	0.0147
	RMSE	1.9712e+02	7.4246e+01	1.0432e-01

4. Conclusion

Conclusion

The mechanical pre-treatment on a Hollander beater of *P. canaliculata* macroalgae is studied. The effect of feedstock/inoculum ratio and the beating time on the resulting methane yields is evaluated. An accurate fuzzy logic (FL) model to estimate the methane yield based experimental datasets is proposed. To achieve an acceptable testing error, the fuzzy model using 2 epochs has been trained until the target is met. The predicted data using the fuzzy model is dispersed nearby to the diagonal line, which confirms the superiority of the model for prediction purposes. The coefficient of determination values are 0.99775 and 0.99074, respectively, for FL and Response Surface Methodology. But the RMSE values are 3.8122 and 8.1294 respectively for FL and Response Surface Methodology. This demonstrated the high precision of FL modeling compared with ANOVA. Then, to identify the optimal operating conditions of the process, a PSO technique has been utilized. Two input parameters; beating time and feedstock/inoculum ratio are assigned as decision variables during the optimization procedure for maximization of methane yield. The obtained optimized results endorse the superiority of the integration of Fuzzy logic and PSO compared with ANOVA. The best values of methane yields are obtained for high pre-treatment times and low feedstock to inoculum ratios, allowing better exploitation of the macroalgae biomass. However, according to this study, a value of 340.1674 ml/gVS of methane yield can be reached at a beating time of 60 minutes and an F/I ratio of 0.3096.

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