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Research Article

Ultrasonic assisted extraction of biooil from *Ricinus communis* seeds: Optimization using Response Surface Methodology and Artificial Neural Network

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Abstract

Currently, there is high energy demand due to fast depletion of fossil fuel resources. Biofuels are the best alternative sources to meet the future energy demand. The present work was focused on optimization of ultrasonic assisted extraction of biooil from *Ricinus communis* seeds using Response Surface Methodology (RSM) and Artificial Neural Network (ANN) linked Genetic Algorithm (GA). Extraction parameters such as solvent concentration, mass of seed, extraction time and temperature were optimized under the constant influence of ultrasonic bath at 50 KHz. The RSM and ANN predicted biooil yields were compared to the experimental yield. The ANN model was found more efficient than RSM. The maximum biooil yield of 55.23% was obtained for optimal solvent concentration of 8 (v/w), mass of seed of 4 g, extraction time of 40 min at 40°C under the ultrasonic bath.

Keywords: Ultrasonic extraction; *Ricinus communis* seeds; Biooil; Optimization; Artificial Neural Network.

Introduction

Vegetable oils are becoming a promising alternative to diesel fuel because they are renewable in nature. Several non-edible vegetable oils, such as oils from *Pongamia pinnata*, *Jatropha curcas*, *Brassica napus* (Canola) and *Madhuca indica* have been reported as suitable for biodiesel production. The oil yield from the crop itself is always the key factor in deciding the suitability of a feedstock for biodiesel production. The oil yield from the crop itself is always the key factor in deciding the suitability of a feedstock for biodiesel production. Oil crops with higher oil yields are more preferable in the biodiesel industry because they can reduce the production cost [1-3].

Castor plant (*Ricinus communis*) can be cultivated naturally over a wide range of geographical regions under a variety of physical and climatic regimes in all temperate countries of the world. Castor is amongst the plants with the highest oil yield potential because of its high yield of seeds with high oil content. Castor oil is a less expensive vegetable oils, can be used as feedstock in the production of biodiesel. Biodiesel from castor oil is superior for cold winters, because of its exceptionally low cloud and pour points [4]. Castor plant can yield a maximum of 2000 kg oil/ha, whereas rapeseed produces about 1000 kg oil/ha and soybean produces about 500 kg oil/ha. Therefore, castor oil is a promising source to produce biodiesel using less cultivation land [5].

Castor oil is a colourless to very pale yellow liquid with a distinct taste and odour. Boiling point of castor oil is 313°C and its density is 961 kg/m^3 [6]. Castor oil is a triglyceride with a composition of 80-90 % Ricinoleic acid, 3-6 % linoleic acid, 2-4 % oleic acid and 1-5 % saturated fatty acids. Ricinoleic acid is the main fatty acid in castor oil, this fatty acid possesses 18 carbons with three highly reactive functional groups. This feature causes castor oil properties are different from other vegetable oils. The high content of ricinoleic acid with a hydroxyl group is the reason for its high viscosity and density. Castor oil is also

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characterized by its high stability, high hygroscopicity and its solubility in alcohol, which affects the transesterification reaction [7-9]. High solubility of castor oil in methanol favours the transesterification of castor oil into biodiesel [10].

India, China and Brazil have huge export potential for castor oil. Currently mechanical pressing and solvent extraction are used for castor oil production. Mechanical methods are less efficient because of loss of oil into cakes. Solvent extraction using Soxhlet extractor is time consuming and requires high heat energy [8]. It is important to develop efficient extraction process for maximum biooil yield with acceptable purity from castor seeds. Response surface methodology (RSM) is a statistical technique used for developing, improving, and optimizing processes in which responses of interest are influenced by several process variables. Artificial neural network (ANN) is a hidden modelling tool inspired by biological neural networks, which consists of interconnected groups of artificial neurons (input, hidden and output), and processes information using a connectionist approach to computation [11]. Thus the present work was focused on optimization of ultrasonic assisted solvent extraction of biooil from castor seeds using RSM and ANN linked Genetic Algorithm (GA).

Materials and Methods

Materials

Castor (*Ricinus communis*) seeds were collected the local area, Chennai. Chemicals required for extraction such as diethyl ether, ethanol, potassium chloride and hydrochloric acid were purchased from SD fine chemicals, Mumbai, India. The chemicals were of analytical grade and used as received for the experiments without any further purification.

Preparation of castor seeds

The castor seeds was collected and washed to remove the dust. Initially the seeds were dried under sunlight. Then the seeds were again dried in hot air oven to remove the excess moisture over the surface of the seeds. The shell of the castor seeds was removed manually and the kernel was ground to powder using motor and pestle. The finely ground castor seed powder was mixed with desired solvent. Heterogeneous solvent was used for extraction of biooil.

Ultrasonic assisted extraction of biooil from castor seeds

The seed solvent mixture was taken in definite proportions (5 g in 50 ml solvent) in a beaker (100 ml) was completely sealed with aluminum foil to prevent the solvent loss by evaporation. The solvent mixture of diethyl ether:ethanol with 3:1 ratio was used. The ultrasonic bath (model 3.5L 100) operating at a constant frequency of 50 kHz and electric supply 230 V AC was used. The beaker with the seed-solvent mixture was kept inside the ultrasonic bath and system was switched on. After the desired extraction time the ultrasonicated sample was poured into a sterile centrifuge tube and centrifuged at 3500 rpm for 20 min [12].

Biooil separation

Salt or acid treatment is an important pretreatment step in separation of biooil from the solvent mixture. The centrifuged sample consists of two main layers (1) an upper liquid layer that consists of solvent and biooil (2) a lower solid seed cake layer. The aqueous layer was completely separated and subjected to pretreatment. Hydrochloric acid of concentration 38% pure or potassium chloride (Analytical grade) was added at constant dilutions to the aqueous layer in the ratio of 1:5 (v:v) for pretreatment of liquid layer. The pretreatment of liquid phase results in two layers; the upper layer comprises of biooil and lower transparent layer consists of excess solvents. The biooil was carefully separated using a separating funnel and stored for further analysis. The weight of biooil was measured and the percentage of biooil yield (w/w) was calculated.

Optimization of extraction conditions using response surface methodology

RSM is a statistical optimization technique, based on the fundamental principles of statistics, randomization, replication and, duplication, which simplifies the optimization by studying the mutual interactions among the variables over a range of values in a statistically valid manner. The three level central composite design (CCD) of RSM consist of 31 experiments was developed for the optimization of solvent concentration, mass of seed, extraction time and temperature under constant influence of ultrasonic bath at 50 KHz for biooil extraction using reflux condenser. The low level (-1) and high level (+1) of extraction parameters are, solvent concentration 5-15% (v/w), mass of seed from 1-3 g, extraction time 20-60 min and temperature 30-50°C respectively. The CCD in actual unit of the 4 variables (Table 1) was developed using Minitab 15 software. Experimental results of CCD were fit into full second-order polynomial model according to eq. (1).

$$Y = b_0 + \sum b_i X_i + \sum b_{ii} X_i^2 + \sum b_{ij} X_i X_j \qquad (1)$$

Where Y is the response variable to be modelled, X_i and X_j are independent variables in coded units and b_i, b_{ii}, b_{ij} are the measures of the X_i, X_j, X_i^2 and X_iX_j of linear, quadratic and interaction effects respectively [13, 14]. The second order polynomial model was maximized by a constraint search procedure using the MINITAB software 15 to obtain the optimal levels of the independent variables and the predicted maximum biooil yield.

Optimization of extraction conditions using artificial neural network linked genetic algorithm

A sigmoidal transfer function was adopted in this study. The input vector (X) was considered to be the solvent concentration (X_1), mass of the seed (X_2), extraction time (X_3), and ultrasonic bath temperature (X_4). Whereas the output vector was the percentage of biooil yield (Y). Once the ANN model was developed and listed, GA was adopted to optimize the model at particular range based on the evolutionary methods. The individual represents the set of independent variables for the evaluation of its fitness of an objective function [15, 16]. The training was carried out by adjusting the connection weights and biases between neurons with the aim of reducing the mean square error (MSE) between the predicted and experimental outputs below an acceptable threshold. The validated model was subsequently used as an evaluation function for the GA optimization process to predict the optimal values of extraction variables (X_1, X_2, X_3, X_4 and Y) applied in developing the model [11, 17, 18].

Results and discussions

Optimization of extraction conditions using response surface methodology

The CCD for four process parameters namely solvent concentration (X_1), mass of seeds (X_2), extraction time (X_3) and temperature (X_4) experimental and predicted yield of biooil is given in table 1. The student t-test was used to analyse individual, square and interaction effects of extraction conditions on biooil yield using experimental results obtained using CCD design in table 2.

The biooil yield is directly fitted to second order polynomial model as shown in eq. (2) using the estimated regression co-efficients given in table 2.

$$Y=43.934-0.59X_1+3.435X_2+2.906X_3+3.147X_4 - 15.482X_1 \times X_1 + 10.143X_2 \times X_2 + 0.103X_3 \times X_3 - 2.817X_4 \qquad (2)$$

Where Y is the % of biooil yield and X_1, X_2, X_3, X_4 are the independent variables in coded units. It was observed that the overall effect of extraction conditions significantly (P<0.001) increased the biooil yield as shown in table 2. The extraction conditions such as solvent concentration, mass of seed showed greater influence from the low level (-) to medium level (0) whereas the operating parameters such as solvent concentration, mass of seed and temperature from medium level (0) to high level (+1) showed a decrease in biooil yield (P<0.001).

The quadratic effects of the operating parameters showed no significant influence (P>0.01) on the % of biooil yield. The CCD of the predicted was similar to the experimental yield of the biooil as shown in table 1. The statistical significance of the quadratic regression model, the individual factors, their interactions and the goodness of fit by analysis of variance (ANOVA) are shown in shown in table 3. The ANOVA of the polynomial model demonstrates that the model is significant (P>0.01).

The interaction between the input variables on biooil yield is graphically shown as contour plots in fig. 1. The shape of contour plots indicates the significant interaction between the each variable showing the circular or ellipse. Contour plots shows the intensity of the interaction between any two independent variables by considering other variables as constant. The central ellipse in the contour plot represents the highest predicted value for the response value. It can be observed from the fig. 1 that the increase in solvent concentration has increased the biooil yield gradually. The extraction time and the temperature have shown significant interaction on biooil yield with the maximum temperature of 50°C. The influence of mass of seed can be observed from the contour plot showing its significant interaction with other input variables. The significant interaction of temperature and solvent concentration on biooil yield was also observed from fig. 1. The maximum biooil yield of 54.97% was predicted for solvent concentration of 10 (v/w), mass of seed (2.15 g), in extraction time of 60 min at 50°C under the ultrasonic bath. The regression model developed using RSM was accurate in prediction of biooil yield which is evident from the R^2 value of 0.962.

Table 1. Three level CCD in actual units for optimization of ultrasonic assisted biooil extraction from castor seeds

Std. Exp. Order	Extraction Conditions				Biooil yield (Y), % (w/w)		
	Solvent, ml/g	Seed, g	Time, min	Temperature, °C	Experimental	Predicted (RSM)	Predicted (ANN)
1	5	2	20	30	20.70	21.29	20.99
2	15	2	20	30	24.50	25.45	24.50
3	5	4	20	30	45.10	43.80	45.10
4	15	4	20	30	41.00	41.23	41.00
5	5	2	60	30	21.00	24.54	20.71
6	15	2	60	30	36.95	33.00	36.94
7	5	4	60	30	37.93	35.41	37.94
8	15	4	60	30	34.85	37.15	34.86
9	5	2	20	50	32.80	31.94	32.80
10	15	2	20	50	26.65	27.84	26.64
11	5	4	20	50	38.90	41.53	38.90
12	15	4	20	50	32.80	30.71	32.81
13	5	2	60	50	49.20	47.65	49.20
14	15	2	60	50	45.10	47.85	45.11
15	5	4	60	50	45.10	45.60	45.12
16	15	4	60	50	41.00	39.09	41.01
17	5	3	40	40	30.07	29.04	30.04
18	15	3	40	40	27.33	27.86	27.32
19	10	2	40	40	53.30	50.64	53.34
20	10	4	40	40	55.35	57.51	54.80
21	10	3	20	40	42.47	41.13	42.49
22	10	3	60	40	46.10	46.94	46.08
23	10	3	40	30	37.80	37.97	37.80
24	10	3	40	50	44.93	44.26	44.91
25	10	3	40	40	43.79	43.93	43.72
26	10	3	40	40	43.73	43.93	43.72
27	10	3	40	40	43.67	43.93	43.72
28	10	3	40	40	43.63	43.93	43.72
29	10	3	40	40	43.75	43.93	43.72
30	10	3	40	40	43.76	43.93	43.72
31	10	3	40	40	43.72	43.93	43.72

Optimization of extraction conditions using artificial neural network linked genetic algorithm

Multilayer normal feed forward neural network was used to train and test the experimental results obtained using CCD (Table 1). The modelling of ANN consists of an input layer with four neurons namely solvent concentration, mass of seed, extraction time and temperature with the hidden and the output layer (biooil yield). The feed forward neural network was trained using incremental back propagation (IBP) algorithm. All neurons from the hidden layer and output layer were calculated by applying a sigmoidal transfer function to avoid overtraining and to decrease training time. The predications of ANN model was more accurate than the RSM based regression model which are identified by the coefficient of determination (R^2 value).

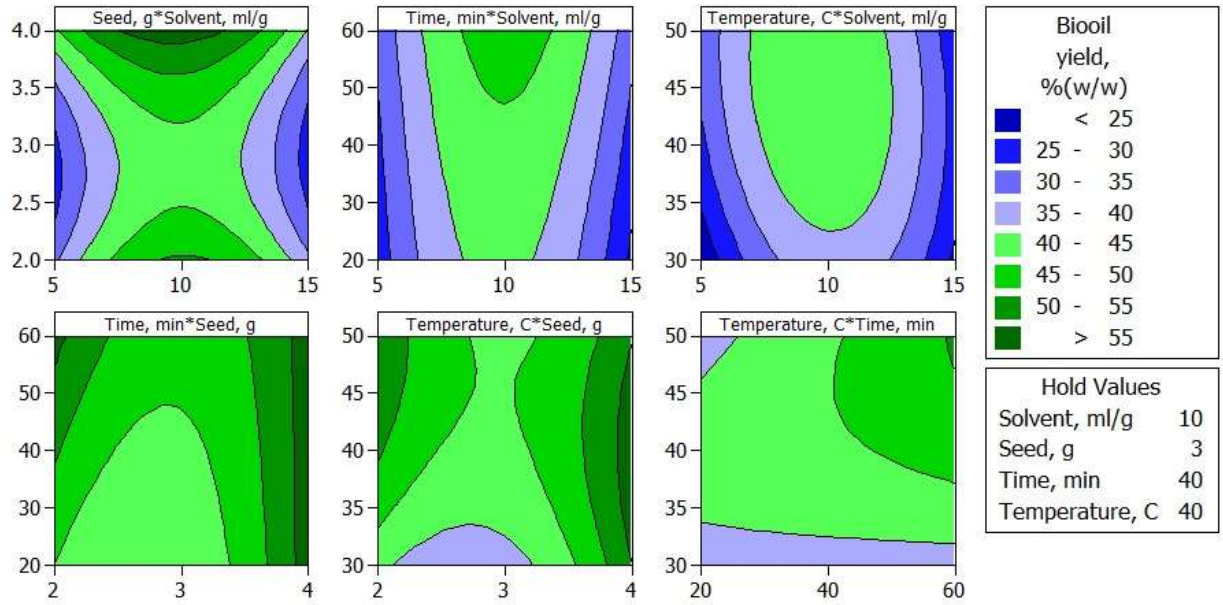

Fig. 1. Interaction effect of various input variables on biooil yield

Table 2. Estimated coefficient of determination for optimization of ultrasonic assisted biooil extraction from castor seeds

Term	Coef	SE Coef	T	P	Significance
Constant	43.934	0.689	63.695	<0.001	Significant
X_1	-0.590	0.548	-1.077	0.298	Not significant
X_2	3.435	0.548	6.268	<0.001	Significant
X_3	2.906	0.548	5.303	<0.001	Significant
X_4	3.147	0.548	5.743	<0.001	Significant
X_1*X_1	-15.482	1.443	-10.727	<0.001	Significant
X_2*X_2	10.143	1.443	7.027	<0.001	Significant
X_3*X_3	0.103	1.443	0.071	0.944	Not significant
X_4*X_4	-2.817	1.443	-1.952	0.069	Not significant
X_1*X_2	-1.680	0.581	-2.890	0.011	Significant
X_1*X_3	1.076	0.581	1.851	0.083	Not significant
X_1*X_4	-2.064	0.581	-3.550	0.003	Significant
X_2*X_3	-2.908	0.581	-5.002	<0.001	Significant
X_2*X_4	-3.230	0.581	-5.557	<0.001	Significant
X_3*X_4	3.114	0.581	5.357	<0.001	Significant
R^2 = 96.2%; R^2(adj) = 92.9%.					

The scatter plot shown in fig. 2 shows the proximity of experimental and predicted biooil yield RSM and ANN. The predicted coefficient of determination of ANN model ($R^{2=}0.999$) was higher than the RSM regression model ($R^{2=}0.961$). Once the ANN model was trained and tested, the GA was applied to find the optimal extraction conditions for maximum biooil yield. The predicted ANN model was optimized using GA with population size of 30, a mutation rate of 0.1 and a uniform crossover rate of 0.8 using random selection for the optimization of input variables for maximum biooil yield.

Table 3. ANOVA for optimization of ultrasonic assisted biooil extraction from castor seeds

Source	Degree of Freedom	Seq Sum of Squares	Adj Sum of Squares	Adj Mean Square	F-value	p-value
Regression	14	2199.10	2199.10	157.078	29.05	<0.001
Linear	4	548.96	548.96	137.240	25.38	<0.001
Square	4	1060.99	1060.99	265.248	49.06	<0.001
Interaction	6	589.15	589.15	98.191	18.16	<0.001
Residual Error	16	86.50	86.50	5.406		
Lack-of-Fit	10	86.49	86.49	8.649	2869.20	<0.001
Pure Error	6	0.02	0.02	0.003		
Total	30	2285.60				

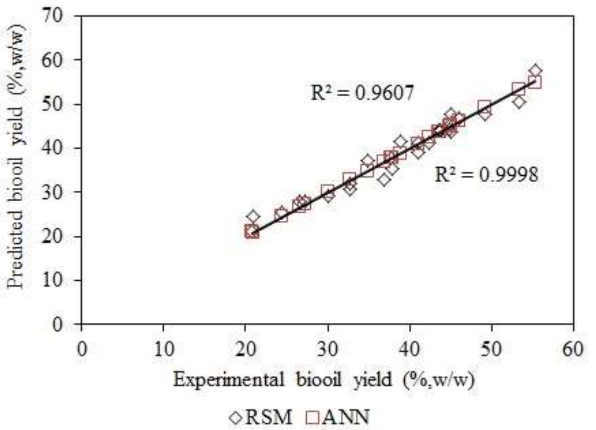

Fig. 2. Experimental versus RSM and ANN Predicted biooil yield

The percentage importance of extraction conditions on biooil yield was identified by using GA as shown in fig. 3. The relative significance of solvent concentration was 33%, mass of the seed was 28% followed by extraction time 27% and temperature 12% as shown in fig. 3. The ANN linked GA model predicted maximum biooil yield was 55.23% using solvent concentration 8 (v/w), mass of seed 4 g, extraction time 40 min and temperature 40°C with R^2 value of 0.999. The relative correlation data on the biooil yield of predicted value was similar to the experimental value. The coefficient of determination for predicted biooil obtained is by ANN model was more closer to 1 than RSM based regression model. Thus the ANN model is more efficient in prediction of biooil yield than RSM model. An experimental biooil yield of 55.97% was obtained using ANN predicted optimal conditions, indicates ANN model is more accurate in prediction of optimal conditions for ultrasonic assisted biooil extraction from castror seeds. Though percentage yield of biooil by both RSM and ANN are very closer, the more amounts of castor seeds can be processed using less solvent, in less time and temperature using ANN optimized conditions. Thus ANN linked GA is more efficient than RSM for optimization of ultrasonic assisted extraction of biooil from castor seeds.

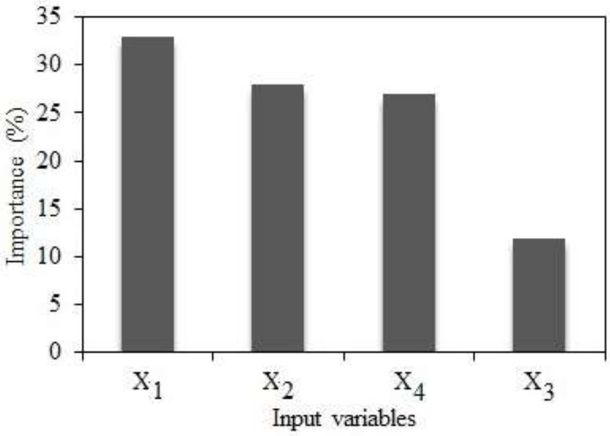

Fig. 3. Percentage importance of input variables on biooil yield

Conclusions

The extraction of biooil from castor seeds was found to be efficiently optimized using both RSM and ANN linked GA. Both the methods are effective in describing the parametric effect of the extraction conditions on biooil yield. The coefficient of determination was used to test the efficiency of RSM and ANN models. The ANN model was found more accurate with higher coefficient of determination than RSM model, with a low percentage relative error (0.005%). The biooil yield of 55.97% was obtained using optimal conditions of ANN linked GA. Thus ANN linked GA was found to be more effective than the RSM for ultrasonic assisted biooil extraction from castor seeds.

Conflicts of interest

The authors declare no conflict of interest.

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