

Optimization of Vacuum Pyrolysis Process Using Generalized Regression Neural Network

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Abstract

A new empirical technique to construct predictive models of vacuum pyrolysis process is presented in this study. Pyrolysis of biomass for preparing bio-oil was studied on a vacuum pyrolysis system, where rape straw was chosen as the raw material. The experiments ran based on orthogonal experimental design method. The operation factors of the system including pyrolysis temperature, system pressure, heating rate and holding time were chosen as input variables, while bio-oil yield and energy transformation ratio were selected as output to establish the prediction models based on Generalized Regression Neural Network (GRNN). The operation factors of the system were optimized for maximizing bio-oil yield and energy transformation ratio, and the optimization result was confirmed by experiments. The results of research showed that the predicted values are fit well with the experimental values, which verifies the effectiveness of the prediction models. Optimal conditions are obtained at pyrolysis temperature of 486.8 °C, heating rate of 18.1 °C/min, reactor pressure of 5.0kPa and holding time of 55.0min. Confirmation runs give 41.9%, 42.5% and 42.1% of bio-oil yield and 34.3%, 34.0% and 34.9% of energy transformation ratio compared to 43.6% and 35.5% of predicted value. Therefore, the forecasting model based on the GRNN is able to result in good prediction and has research value to the reality.

Keywords: vacuum pyrolysis, bio-oil yield, energy transformation ratio, optimization, GRNN

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1. Introduction

With the continuous development of modernization process of society, the contradiction between supply and demand of energy will become increasingly prominent. According to the Statistics data released by National Bureau of China in February 2013, crude oil imports in 2012 reached 270 million tons, accounting for 60% crude oil consumption of the whole year [1]. Social production and energy security situation of China become more serious, and long-term use of petroleum energy causes a serious negative impact on the environment, such as the greenhouse effect, global warming, destruction of the ecological balance and acid rain. The total theory national biomass resources are about 50 billion tons of standard coal, which is nearly 1.5 times of the total energy consumption in China [2]. So it is significant to ease the pressure of energy and environment by making full use of biomass resources. The biomass with relatively low energy density can be converted to liquid fuel with higher energy density by pyrolysis liquefaction, which makes biomass pyrolysis liquefaction becoming a hotspot of alternative energy research around the world [3-5]. Biomass pyrolysis can be divided into several types according to the reaction conditions [6], such as conventional pyrolysis, fast pyrolysis, vacuum pyrolysis and so on. The vacuum pyrolysis attracts extensive attention from researchers, whose liquefied energy consumption is relatively low and bio-oil yield is high [7, 8]. The bio-oil yield and energy transformation ratio are affected by pyrolysis system operation factors and raw material properties [9], and it is a nonlinear relationship between these parameters and evaluation index of pyrolytic liquefaction, which makes the prediction of evaluation index become more complicated. The complexity has been circumvented in an empirical way [10]. Over the years, many scholars have tried a variety of traditional methods to predict, such as regression model, grey prediction model, Markov model and so on [10-12]. Although current forecasting methods have advantages, the established model can not essence and comprehensively reflect the inner structure and complex characteristics of the prediction dynamic data, which can lost a certain amount of information.

While artificial neural network (ANN) has unique characteristics, such as parallel

structure, adaptive, self-organization, associative memory, strong fault tolerance and robustness, and unique information processing method, adapted the complexity, nonlinearity and uncertainty, and made remarkable achievements in practical [10, 13]. Radial basis function (RBF) networks, especially Generalized Regression Neural Network (GRNN), is able to serve as a predictor for estimating the future values of the variables or for modeling the process or system of output variables to input variables. GRNN provides estimates of continuous variables and converges to the underlying (linear or nonlinear) regression surface. This generalized regression neural network is a one-pass learning algorithm with a highly parallel structure. The algorithm form can be used for any regression problem in which an assumption of linearity is not justified. One of the impressive advantages of GRNN can be designed very quickly. It has property of requiring no iterative training. It approximates a function between input and output vectors, drawing the function directly from the training data. Furthermore, it is consistent. If the training set size becomes large, the estimation error approach zero. The GRNN has achieved good results in the prediction study [14, 15].

Currently, the use of ANN simulation prediction is applied to biomass pyrolysis gasification and the pyrolysis weightlessness process [16, 17]. In this study, multi-factor and multi-level orthogonal experiments for pyrolysis liquefaction were presented, where rape straw was chosen as the raw material. With pyrolysis system factors as input, the GRNN prediction models of bio-oil yield and energy transformation ratio were established and targeted to system factor optimization about bio-oil yield and energy transformation ratio, which provided experimental and theoretical basis for the high-efficient conversion and utilization of biomass resources.

2. Experiment and Methodology

2.1. Materials

The rape straw used in this study was collected from rural area in Zhenjiang, China. The Rape straw was ground and sieved for small particles in the 100~150 μ m range. Lu et al. [18] found that a decrease in particle size will tend to higher liquid product yields. The sample was dried at 105 $^{\circ}$ C for 2h to remove external moisture. The proximate analysis was performed according to ASTM D-2974 using the thermo-gravimetric analysis TGA/DSC 1 for determination of moisture, volatile matter, fixed carbon and ash content in rape straw. The ultimate analysis of the dried sample was carried out in an elemental analyzer (Model: FLASH1112A, Italy). The results of proximate, ultimate analysis of rape straw are shown in Table 1.

Table 1. Proximate and Ultimate Analysis of Rape Straw

M _{ad}	A _{ad}	V _{ad}	FC _{ad}	C	H	N	S	O ^a
6.12%	3.69%	72.84%	17.35%	42.22%	5.53%	0.41%	0.07%	51.77%

_{ad}, Air dry base; ^aOxygen by difference.

2.2. Experimental Procedure

Vacuum pyrolysis of rape straw was conducted in a fixed-bed reactor. The scheme of the vacuum pyrolysis system is shown in Figure 1. The reactor consisted of a 1.5m long quartz tube (diameter 60mm), heated by six well-insulated, computer controlled heating elements. The heated reactor chamber was connected to a condensation trap and a vacuum pump. The pipes leading from the reactor to the condensation trap were maintained at 150 $^{\circ}$ C to limit condensation before the trap. The condensation temperature was set at -20 $^{\circ}$ C for all experiments (The cooling medium: ethylene glycol). The bio-oil collected was then removed and weighed. A control program was used to control the final pyrolysis temperature, heating rate and pyrolysis time. Once the reactor was loaded (150 \pm 5g of biomass), heating started after the reactor was sealed and evacuated to the desired operating pressure. A typical run would take between 1h and 2h depending on the conditions employed. At last, Protective gas (Nitrogen) was introduced into the reactor to prevent products oxidation at high temperature. Pyrolysis experiments were run according to orthogonal design.

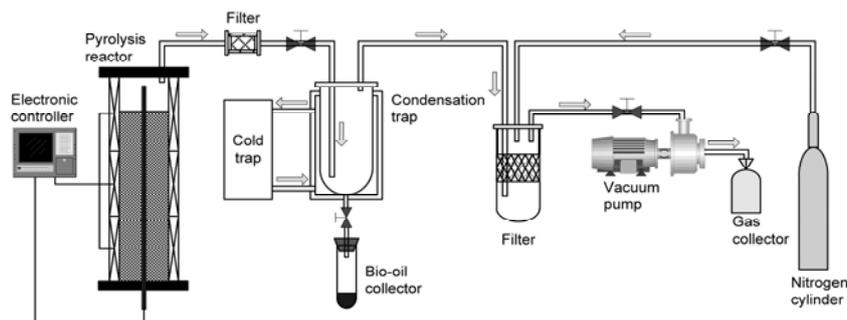


Figure 1. Scheme of the Vacuum Pyrolysis System

2.3. Experimental Design

In this study, four main factors that have effects on bio-oil yield were considered, including pyrolysis temperature, reactor pressure, heating rate and holding time. As shown in Table 2, each factor has five levels in the range of experimental parameters. For this problem, the minimum orthogonal array is selected as $L_{25}(5^4)$. All experimental runs were repeated to ensure reliability of the data and results' reproducibility. Experiment variables and levels are shown in Table 2.

Table 2. Experiment Variables and Levels

Levels	Pyrolysis temperature $x_1/^\circ\text{C}$	System pressure x_2/kPa	Heating rate $x_3/^\circ\text{C}\cdot\text{min}^{-1}$	Holding time x_4/min
-2	400	5	4	15
-1	450	20	8	30
0	500	35	12	45
1	550	50	16	60
2	600	65	20	75

2.4. Calculation Method

In the results presented, the yield of the bio-oil has been defined as Equation (1):

$$\text{Bio-oil yield} = \frac{m_l}{m} \times 100\% \quad (1)$$

Where m is the initial mass of rape straw [g], m_l is the mass of collected bio-oil [g].

The high heat value (HHV) of the bio-oil was measured by using a bomb calorimeter, so energy transformation ratio of vacuum pyrolysis liquefaction process can be obtained as Equation (2):

$$\text{Energy transformation ratio} = \frac{q_l m_l}{qm} \times 100\% \quad (2)$$

Where q is the HHV of rape straw [17883kJ/kg], and q_l is the HHV of the bio-oil [kJ/kg].

2.5. Principle of Generalized Regression Neural Network

The joint probability density function $f(x, y)$ is assumed to be the random vector x and the random variable y . When observation vector of x is x_0 , the expected value of y relative to x_0 regression is:

$$Y = E[y | x_0] = \frac{\int_{-\infty}^{\infty} y f(x_0, y) dy}{\int_{-\infty}^{\infty} f(x_0, y) dy} \quad (3)$$

The unknown probability density function $f(x_0, y)$ can be obtained by training samples $\{x_i, y_i\}_i^n$ through the Parzen nonparametric estimates [18]:

$$\hat{f}(x_0, y) = \frac{1}{(2\pi)^{\frac{p+1}{2}} \sigma^{(p+1)} n} \sum_{i=1}^n e^{-\frac{D_i^2}{2\sigma^2}} e^{-\frac{(y-y_i)^2}{2\sigma^2}} \tag{4}$$

Where p is the dimension of the input vector; n is training sample capacity; σ is the smooth factor; D_i is euclidean distance between x_0 and x_i . The formula is defined an Equation (5):

$$D_i = \|x_0 - x_i\| = \sqrt{\sum_{j=1}^p (x_{0,j} - x_{i,j})^2} \tag{5}$$

$f(x_0, y)$ is substituted for $\hat{f}(x_0, y)$:

$$Y(x_0) = \frac{\sum_{i=1}^n y_i e^{-\frac{D_i^2}{2\sigma^2}}}{\sum_{i=1}^n e^{-\frac{D_i^2}{2\sigma^2}}} \tag{6}$$

As shown in formula (6), the larger σ is, the greater the input region of the probability density function overwrites is, and the smaller σ is, steeper and closer to the expected value of the network output the probability density function curve is.

The structure of GRNN is shown in Figure 2. GRNN is mainly composed of radial basis neuron layer and a special linear layer.

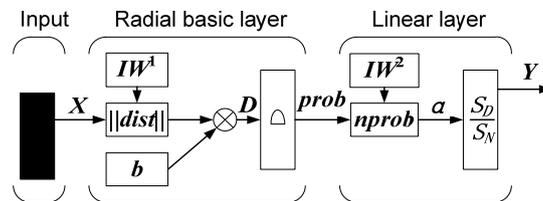


Figure 2. Scheme of Generalized Regression Neural Network

Radial basic layer input is that the Euclidean distance between the input vector X and the neuron weights IW^1 multiplied by the threshold value b , where the neuron weights IW^1 is training sample vector. Output of radial basis neuron layer is defined as Equation (7).

$$prob = \sum_{i=1}^n e^{-\frac{D_i^2}{2\sigma^2}} \tag{7}$$

Firstly, the linear layer nprod module has radial basis neuron layer output multiplied by corresponding variable y_i , and then makes weighted sum, which is defined as Equation (8).

$$S_D = \sum_{i=1}^n y_i prob_i \tag{8}$$

Secondary, the output results are arithmetic summed, which is defined as Equation (9).

$$S_N = \sum_{i=1}^n prob_i \quad (9)$$

The input of the linear transfer function, Y is obtained by the results of S_D and S_N dividing, which is output after processing by the function Y .

3. Results and Discussion

3.1. Experimental Results

Biomass vacuum pyrolysis liquefaction experiments were run according to orthogonal design, including a total of 25 experimental points, and 3 sample test points. Factor combination and experimental results are shown in Table 3.

Table 3. Factor Combination and Experimental Results

Experimental No.	Factor assigned				Bio-oil yield/%	Energy transformation ratio/%
	X ₁	X ₂	X ₃	X ₄		
1	-2	-2	-2	-2	30.40	20.45
2	-2	-1	2	0	37.44	29.85
3	-2	0	1	2	36.61	29.65
4	-2	1	0	-1	34.24	26.45
5	-2	2	-1	1	30.98	22.55
6	-1	-2	2	2	42.30	33.67
7	-1	-1	1	-1	40.98	32.68
8	-1	0	0	1	38.51	30.51
9	-1	1	-1	-2	33.95	24.63
10	-1	2	-2	0	29.04	18.76
11	0	-2	1	1	43.03	34.34
12	0	-1	0	-2	39.52	30.67
13	0	0	-1	0	35.40	26.53
14	0	1	-2	2	29.44	19.31
15	0	2	2	-1	40.47	29.96
16	1	-2	0	0	38.76	30.17
17	1	-1	-1	2	33.59	24.67
18	1	0	-2	-1	27.29	17.70
19	1	1	2	1	39.60	29.05
20	1	2	1	-2	37.70	27.05
21	2	-2	-1	-1	29.49	21.15
22	2	-1	-2	1	22.67	13.69
23	2	0	2	-2	35.96	24.38
24	2	1	1	0	34.02	23.97
25	2	2	0	2	30.23	20.50
26	1	0	0	0	36.69	25.84
27	0	0	1	0	41.04	29.09
28	0	-1	0	0	39.91	30.90

3.2. Establishment of GRNN Prediction Model

(1) Selection of input and output

The samples of number from 1 to 25 were selected as a training set of the network, while number from 26 to 28 are selected as a set of test. While the level value of the four factors about pyrolysis temperature, system pressure, heating rate and holding time setted as the independent variable. For the process of biomass vacuum pyrolysis liquefaction, the ideal situation is that higher bio-oil yields and energy transformation ratio of the bio-oil could be obtained at the same time under a certain condition. Therefore, three GRNN models of different relationships are established.

(2) Experimental data processing

a. Data packet: let the twenty-five (No.1~25) samples as the testing sample and selected the last three (No.26~28) samples as the training sample. b. Data transfer: processed the data of input and output between the interval (0~1), which also known as data normalization. Generally, the data were normalized following formula Equation (10), and then analyzing.

$$\hat{x} = \frac{x - x_{min}}{x_{max} - x_{min}} \quad (10)$$

Where \hat{x} is the data after normalization, x is the true value before normalization, x_{min} is the minimum value of the experimental data, x_{max} is the maximum value of the experimental data.

(3) Establishment of the network

Using the neural network toolbox of the software matlab7.0 programming and constructing the GRNN prediction model. The call format of GRNN is defined as Equation (11):

$$\text{net}=\text{newgrnn}(\text{P},\text{T},\text{SPREAD}) \quad (11)$$

Where P is an $R \times Q$ matrix, T is an $S \times Q$ matrix, SPREAD is the expansion speed of radial basis function, which is the smooth factor, default value is 1. As the smooth factor affecting the performance of the network, the training of GRNN is to find the optimal smooth factor. Therefore, we need to continuously determining the optimal value. Every time we started from 0.1 and increase a unit volume (0.1), obtained the predictive value of the estimated point respectively, and selected the smooth factor as the optimal value, which the corresponded root meansquare error (RMSE) between the predictive of the estimated point and the actual value of the sample is minimum. Model prediction performance was measured by RMSE defined as Equation (12):

$$RSME = \sqrt{\frac{\sum_{j=1}^r (d_j - out_j)^2}{r}} \quad (12)$$

Where d_j and out_j represent actual etch measurement and model prediction corresponding to the j th experiment. The remaining r indicates the total number of test vector. In this study, the method of circuit training was employed and the code as follows:

```

for smooth = 0.1:0.1:2; %% smooth value vassignment
net=newgrnn(p_train,t_train,smooth); %%build GRNN network model
disp(['Current smooth value is', num2str(smooth)]); %%display current smooth
value
test_Out=sim(net,p_cv_test); %%output predicted value under conditions of test
sample
test_Out=postmnmx(test_Out,mint,maxt); %%reverse normalization calculation
error=t_cv_test-test_Out; %%prediction error calculation
disp(['Current mse is',num2str(mse(error))]) %%display current network mean
square error
if mse(error)<mse_max %%determine current mean square error
mse_max=mse(error); %% assignment for mse_max
desired_smooth=smooth; %%assignment for desired_smooth
end
end
disp(['Optimal smooth value is',num2str(desired_smooth)]) %%display optimal smooth
value

```

3.3. Training and Testing

3.3.1. Prediction Model of Bio-oil Yield

According to the cycle validation analysis, when the smooth factor σ_1 was 0.4, the better prediction of bio-oil yield was obtained by GRNN model about system factors and bio-oil yield, which is shown in Table 4 with the comparison of the model prediction values and the experimental values. It can be seen that the absolute value of the relative error between the prediction of the model and the experimental values is less than 5%, indicating that the predicted value is within the acceptable range and the model can be used to predict bio-oil yield.

Table 4. Predictive and Experimental Value of Bio-oil Yield

No.	Experimental values/%	Predicted values/%	Relative error/%
26	36.69	34.97	4.68
27	41.04	40.59	1.10
28	39.91	40.45	-1.35

3.3.2. Prediction Model of Energy Transformation Ratio

According to the cycle validation analysis, when the smooth factor σ_2 was 0.3, the better prediction of energy transformation ratio was obtained by GRNN model about system factors and energy transformation ratio, which is shown in Table 5 with the comparison of the model prediction values and the experimental values. It can be seen that the absolute value of the relative error between the prediction of the model and the experimental values is less than 5%, indicating that the predicted value is within the acceptable range and the model can be used to predict energy transformation ratio.

Table 5. Predictive and Experimental Value of Energy Transformation Ratio

No.	Experimental values/%	Predicted values/%	Relative error/%
26	25.84	26.66	-3.17
27	29.09	30.37	-4.40
28	30.90	31.97	-3.46

3.3.3. Weighted Model of Bio-oil Yield and Energy Transformation Ratio

Assuming that importance of bio-oil yield and energy transformation ratio was equivalent, two each were summed by the weight of 50% of each. When the smooth factor σ_3 was 0.1, GRNN model about the system factors and their weighted values was obtained. The variations with the conditions of the test of bio-oil yield, energy transformation ratio and their weighted values were compared, which is shown in Figure 3. It can be seen that the variations of bio-oil yield, energy transformation ratio and weighted values are similar, indicating that when the predicted value of the weighted model outputs is larger, bio-oil yield and energy transformation ratio can reach a larger value simultaneously, which can be used to optimize the factors of biomass vacuum pyrolysis liquefaction system.

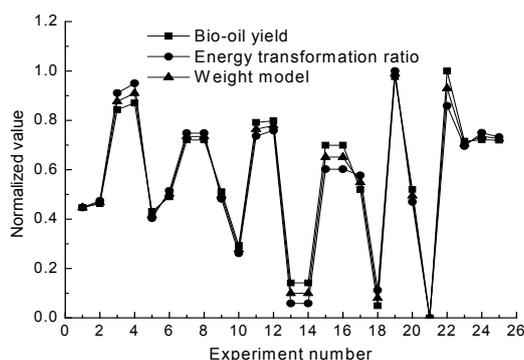


Figure 3. Variations of Three Kinds of Dependent Variables

3.4. Optimization and Confirmation

According to the optimization of the weighted model using Matlab software, when pyrolysis temperature is 486.8°C, system pressure is 5.0kPa, heating rate is 18.1°C/min and holding time is 55.0min, the weighted value is the highest. The conditions are substituted into the GRNN prediction model of bio-oil yield and energy transformation ratio, the predictive values of bio-oil yield and energy transformation ratio are 43.6% and 35.5% respectively. Both of them are higher.

The confirmation runs were performed three times (No.29~31) under this condition to obtain the comparison with predicted values. The predictive and experimental values of bio-oil yield and energy transformation ratio are shown in Table 6. It can be seen that the absolute value of the relative error between the model prediction and the experimental values is less than 5%, indicating that the results of vacuum pyrolysis system factors optimized by GRNN weighted model are accurate, while bio-oil yield and energy transformation ratio obtained under this condition are both higher. Therefore, the forecasting model based on the generalized regression neural network is able to result in good prediction and has research value to the reality.

Table 6. Predictive and Experimental Values of Bio-oil Yield and Energy Transformation Ratio

No.	Bio-oil yield/%			Energy transformation ratio/%		
	Experimental values	Predicted values	Relative error	Experimental values	Predicted values	Relative error
29	41.9	43.6	-4.1	34.3	35.5	-3.5
30	42.5	43.6	-2.6	34.0	35.5	-4.4
31	42.1	43.6	-3.6	34.9	35.5	-1.7

4. Conclusion

In this study, the orthogonal experimental design method was employed to minimize the number of experiments. In further, the GRNN prediction model of bio-oil yield and energy transformation ratio were established using the orthogonal experimental data. The results show that GRNN has a faster learning speed, the ability of dealing with the instability data, accurate prediction for sample, network structure with low human subjective factors and stability prediction, has great practical value for dealing with the complex problems of pyrolysis process. Prediction is accuracy about bio-oil yield and energy transformation ratio. When pyrolysis temperature is 486.8°C, system pressure is 5.0kPa, heating rate of is 18.1°C/min, holding time is 55.0min, bio-oil yield and energy transformation ratio become higher of 43.6% and 35.5% respectively, which is confirmed by the experiments.

Acknowledgements

This work is currently supported by the National Natural Science Foundation of China (No.51276085), the Natural Science Foundation of Jiangsu Province (No.BK2011488) and the Priority Academic Program Development of Jiangsu Higher Education Institutions ([2011]No.6).

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