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Research on Activated Carbon Supercapacitors Electrochemical Properties Based on Improved PSO-BP Neural Network

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Abstract: Supercapacitors, also called electrical double-layer capacitors (EDLCs), occupy a region between batteries and dielectric capacitors on the Ragone plot describing the relation between energy and power. BET specific surface area and specific capacitance are two important electrochemical property parameters for activated carbon EDLCs, which are usually tested by experimental method. However, it is misspent time to repeat lots of experiments for EDLCs' studies. In this investigation, we developed one theoretical model based on improved particle swarm optimization algorithm back propagation (PSO-BP) neural network (NN) to simulate and optimize BET specific surface area and specific capacitance. Comparative studies between the predicted data and experimental data–earlier deduced by Liu et al, have revealed that improved PSO-BPNN model bears higher prediction accuracy, faster computation speed and better generalization performance.It is concluded that the improved PSO–BP NN is one simple and effective method to find optimal conditions of BET specific surface area and specific capacitance for activated carbon EDLCs.

Keywords: Activated Carbon EDLC; Electrochemical Property; Neural Network; Particle Swarm Optimization

1 Intruduction

In recent years, it has aroused lots of interest in supercapacitors (EDLCs) because they fi11 in a gap between batteries and conventional capacitors in terms of energy and power EDLCs have been touted as a solution to the mismatch between the fast

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growth in power required by devices and the inability of batteries to efficiently discharge at high rates [Chmiola, Yushin, Gogotsi, Portet, Simon and Taberna (2006); Arico, Bruce, Scrosati, Tarascon and Schalkwijk (2005)]. This large capacity for high power discharge is directly related to the absence of charge-transfer resistances that are characteristic of battery faradaic reactions and subsequently leads to better performance at low temperature. Improvements in the energy density may accelerate the advent of electrical and fuel-cell cars, as well as enable numerous industrial and consumer applications for supercapacitors [Brodd, Bullock and Leising (2004)]. The main classes of the EDLCs are activated carbon, nano-carbon fiber and carbon aerogel, etc. Among of them, activated carbon has been applied widely because it have many advantages of cheap cost, easy to access and consistent performance.

BET specific surface area and specific capacitance are two important parameters that affect the electrochemical performances (mainly including energy density and power density) of activated carbon EDLCs. During the process of searching for relationship between the technical parameters and desired performance indicators of EDLCs lots of experiments have to be repeated. Such traditional material design method would be bound to waste lots of manpower and resources. However, theoretical modeling offers reasonable alternative because total or part of complex and time-consuming experiments can be replaced[Yang, Gu and Liang (2007); Husain, Guniganti, Sehgal and Pandey (2009)].

Recently, with the development of modern science and technology, artificial intelligent technology has been widely used to predict, estimate and optimize for material engineering[Aymerich and Serra(2006); Kerh, Lai, Gunaratnam and Saunders(2008); Wu, Chiu and Wang (2008)]. BPNN(Back Propagation Neural Network) and PSO (Particle Swarm Optimization) algorithm are two branches of artificial intelligence technology that can be combined to optimize complex non-linear problems [Pidaparti and Neblett(2007)]. In this paper, we proposed a new improved PSO algorithm to optimize electrochemical properties of activated carbon EDLCs. It is found that the improved PSO–BPNN has great potential to handle problems such as estimating and optimizing variables in material chemometrics systems.

2 Experimental process

2.1 Activated Carbon Preparation

A series of activated carbon EDLCs samples, including $A_1 \sim A_{18}$and B_1-B_5 were prepared by means of KOH activation method. The pitch coke powder and KOH solution were mixed with different ratios of alkali / carbon in different heat treatment processes at different activation times. The mixture should be mixed fully,

and then was activated from 700°C to 830°C under nitrogen protection in a nickel autoclave.

2.2 Activated Carbon Electrodes Preparation and Simulation Capacitor Assembly

60£¥ Polytetrafluoroethylene(PTFE) emulsion was added to the electrodes materials as binder and the mass ratio of activated carbon and PTFE was fixed at 95:5. In an agate grinder, the mixture was milled for two hours in order to obtain a homogeneous product. Then, the mixture was kneaded and pressed to shape the round slices electrodes with 13.3mm long and 0.5mm thick by hydraulic machines at 10MPa. The carbon electrode films were made by suppressed carbon film on the nickel foam (100ppi) collector. In an argon glove box, the two equal mass of nickel electrode films were laid face to face and interrupted by polypropylene (PP) septum. Before assembling into simulation capacitors, carbon electrode and PP septum were immersed in a vacuum electrolyte. Finally, they were assembled into a simulated capacitor. The process chart of EDLC electrode was shown in Figure 1.

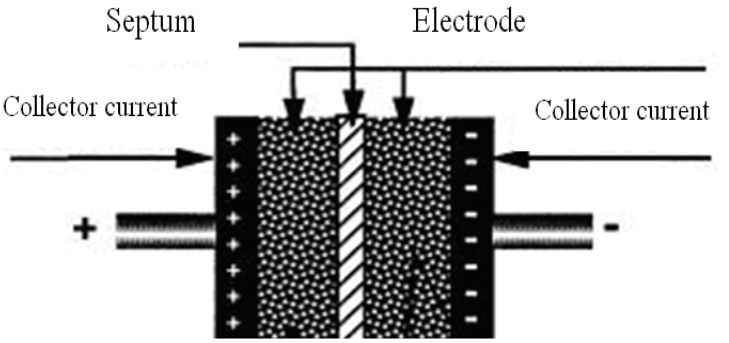

Figure 1: The process chart of EDLC electrode

2.3 Methods of BET Specific Surface area and Specific Capacitance Test

Specific surface area of EDLC electrode was tested by multipoint BET method and the BET specific surface area was measured by Micromeritics ASAP2020.

Constant current charge-discharge circulating was tested with DTS which is a kind of high-precision capacitors test system. The range of charge voltage was 0.05V $\sim$0.9 V and the number of charge and discharge circulating was more than one hundred. We obtained the specific capacitance of activated carbon simulation capacitor

according to [Qu and Shi (1998)]:

$$C = 2I(m \times \Delta V / \Delta t), \tag{1}$$

where $I(A)$ is the discharge current, m(g) is the mass of single carbon electrode and $\Delta V/\Delta t$ is the slope of constant current discharge .

2.4 Experiment Data

BET specific surface area or S_{BET} and specific capacitance of activated carbon EDLCs were determined by lots of factors. In this study, we focused on three key factors: activation dosage, activation time and activation temperature. The practice for producing activated carbon EDLCs showed that selecting appropriate activation dosage, activation time and activation temperature can effectively adjusting and controlling experiment condition, achieve good technical parameters.Technical parameters of experimental samples were listed in Table 1.

3 BP Artificial Neural Networks

Artificial neural network or NN was founded by McCulloch and Coworkers in early 1940s [Perlovsky (2001)]. NNs had emerged as a result of simulation of biological nervous system (such as the brain) on a computer. Nowadays, as one of the most powerful computer analysis techniques, neural networks have been used in many fields. By way of researching the application of nerve network technique, lots of the computer neural network models were established [Gorynin and Ushkov (2000)].

Materials chemometrics involves the relationships among components, process, organization and performance. However, it is difficult to establish mathematical model due to complex and unclear internal laws of material system. To overcome this problem, NNs use samples data to establish material intelligent system models. NNs have many advantages such as strong ability of nonlinear approximation, self-learning and so on. More importantly, NNs have the ability to solve multi-parameter and multi-step prediction problems. Just for that, NNs are very suitable for materials chemometrics problems.

There are dozens of neural network model such as BP, Radial Basis Function or RBF, Kohonen, Hopfield and Generalized Regression Neural Network or GRNN. Among of them, BPNN is the most extensive and practical one. The vast majority of neural networks are rooted in the standardized BP algorithm. Figure 2 showed the typical structure of BPNN[Yuan (2002)]. BP neural network is usually comprised by three layers. The first layer is input layer consisting of a group of processing units which are responsible for acceptance of data imported to the net-

Table 1: Technical parameters of experiment samples

Sample	KOH/ Pitch coke	Activation time /h	Activation temperature/^{o}C	S_{BET} /$m^2.g^{-1}$	Capacitance /$F.g^{-1}$
A1	6: 1	1.5	830	3396	298
A2	5: 1	1.5	830	3134	269
A3	4: 1	1.5	830	2787	238
A4	3: 1	1.5	830	2261	228
A5	5: 1	1.5	800	2532	247
A6	4: 1	1.5	800	2465	236
A7	4.5£º1	1.5	800	2649	248
A8	3: 1	1.5	800	1861	223
A9	4: 1	2	800	2663	269
A10	4: 1	3	800	2963	257
A11	3: 1	4	800	2459	236
A12	3: 1	3	800	1827	219
A13	3: 1	2	700	1750	212
A14	6: 1	1.5	770	3365	287
A15	5: 1	1.5	770	3124	241
A16	4: 1	1.5	770	2598	233
A17	3: 1	1.5	770	1980	222
A18	2: 1	1.5	800	1229	203
B1	6: 1	2	800	3013	284
B2	4: 1	1.5	800	2663	237
B3	4: 1	2	770	2559	226
B4	3: 1	1.5	700	2169	219
B5	5: 1	3	800	2732	245

work. The output layer is responsible for output data to the network. Other layers which do not accept any input data or output data are called hidden layers.

In Figure 2, supposing that w_{ji}^l is connection weight from i node of $l-1$ layer to j node of l layer, $net_j^l$and out_j^l are input and output values of j node of l layer, $X_i(i=1,\cdots N)$is input factor, f is neurons activation function. Usually, BPNN is

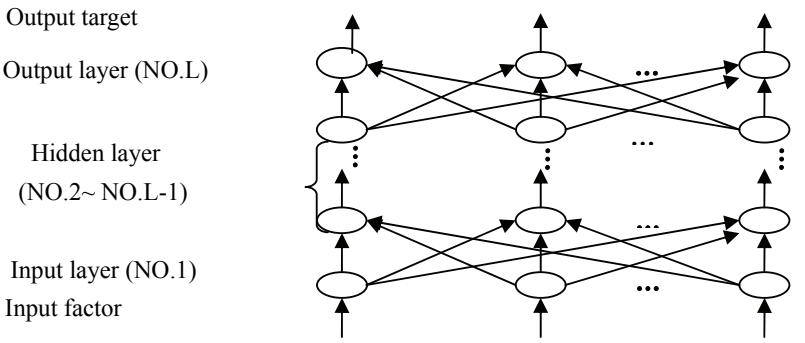

Figure 2: Basic structure of BPNN

represented by following model:

$$out_j^1 = x_j \quad (j = 0,1,\ldots,N) \tag{2}$$

$$net_j^l = \sum_{i=0}^{pot(l-1)} \omega_{ji}^l out_i^{l-1} \quad (l = 2,3,\ldots,L) \tag{3}$$

$$out_j^l = f(net_j^l) \quad (j = 1,2,\ldots,pot(l)) \tag{4}$$

$$\hat{E}_j = out_j^L \quad (j = 1,2,\ldots,M) \tag{5}$$

Here, $pot(l)$ $(l = 1,2,\cdots,L)$ in Eq.(4) is the node number of each layer $\hat{E}_j$ is estimated value of design objective. In BPNN, hidden layers error is determined by back propagating algorithm. BP algorithm is repeated several times until obtaining reasonable residual error. Steps of optimization procedure are similar to those detailed in [Guessasma, Montavon, Gougeon and Coddet (2003)] and the learning algorithm is detailed in [Hamzaoui, Guessasma, ElKedim and Gaffet (2005)].

4 Improved Particle Swarm Optimization Algorithm

The particle swarm optimization algorithm was first put forward by Kennedy and Eberhart[Eberhart and Kennedy (1995)] who was inspired by the swarming behavior of animals and human social behavior. In PSO algorithm, a point in the problem space is called a particle which consists of a position vector$\vec{x}$ and a velocity vector$\vec{v}$. Each particle's position property is treated as a solution in D-dimensional space. Each particle is 'flown' through the multidimensional search space and adjusts its position according to its own experience and those of neighboring particles. The performance of each particle is evaluated by a predefined fitness function.

PSO is initialized by a group of random particles with different positions property and velocities property. The optimal solution is obtained through iteration. The two 'extremes' are the best position $\vec{p}$obtained by the particle so far and the overall best position $\vec{p}_g$ obtained by all particles. Two 'extremes' are updated in each iteration. The updates of each particle consist of velocity update and position update.

Usually, the particle position is updated according to:

$$\vec{x}(t+1) \leftarrow \vec{x}(t) + \vec{v}(t+1), \tag{6}$$

and the velocity is updated according to:

$$\vec{v}(t+1) = \omega\vec{v}(t) + \varphi_1(\vec{p} - \vec{x}(t)) + \varphi_2(\vec{p}_g - \vec{x}(t)), \tag{7}$$

where φ_1 and φ_2 are accelerated coefficient used to adjust the $\vec{v}_g$ and $\vec{v}$ respectively. They are often selected as random numbers typically within[0,2]. w is the nonlinear variation inertia weight that represents speed inertial of particles.

In this work, step expression is replaced by Eq.(8):

$$\vec{x}(t+1) \leftarrow c_1 rand_1 \vec{x}(t) + c_2 rand_2 \vec{v}(t+1), \tag{8}$$

where c_1 and $c_2$are step coefficients which are used to adjust the $\vec{x}_g$ and $\vec{x}$ respectively. To accelerate convergence rates and improve numerical precision,c_1 and c_2 often are selected as 2. $rand_1$and $rand_2$ are random numbers typically within the limits of [-1,1]. In comparing improved variable step length PSO algorithm with the traditional PSO, improved PSO has following advantages: straight-forward logic, easy implement, few parameters to adjust and rapidly convergence rates because the step length is controlled by the autocorrelation estimation of error signal. Improved variable steplength PSO algorithm constantly updates using Eq.(7) and Eq.(8) until specified number of iteration is exceeded or the particle velocity is close to zero.

5 Improved Particle Swarm Optimization Neural Network Model

Gradient algorithm has been widely used for training the weights of feed forward neural networks. BP algorithm has strong ability of searching for local optimal, however it is difficult for BP algorithm to get the global optimization as BP is a kind of gradient-based method. The weight is often updated according to:

$$\omega_{ji}^N(k) = \omega_{ji}^N(k-1) + \Delta\omega_{ji}, \tag{9}$$

where N is the number of input node and K is the number of iteration. As usual, BP algorithm has exposed these shortcomings: slowly convergence training speed,

easily to get stuck into a local minimum and the final weight value depending heavily on the initial weight. As the initial weight is given randomly, this would also heavily affect speed and accuracy if the free given initial value is not ideal. To overcome these problems the PSO algorithm has been adopted.

PSO is one of global algorithm which has strong ability of searching for global optimal. PSO improves the efficiency of search and avoids precocity because its optimization process dose not depend on the gradient information. By introducing the nonlinear variation weight and step coefficients into the standard particle swarm algorithm ensures the overall convergence and enhances the accuracy of convergence. By introducing inertia factor, the movement inertia of particle maintains good momentum. It is helpful to search for the new better regional for the particles. The optimization results of NN would less depend on the random initial particles.

In this paper, improved PSO was used to optimize the initial weight and train threshold of BPNN model. Firstly, using PSO algorithm, optimized input and output weights and threshold of BP model were obtained. Then, after initial value optimizing, the optimized PSOBPNN model was used to predict electrochemical properties of activated carbon EDLCs. Thus, some problems about BPNN such as slowly convergences training peed, easily to get stuck into a local minimum, sensitive initial weight and other issues could be well solved.

6 PSO-BPNN Predicting Model for Activated Carbon EDLC's Electrochemical Properties

The main process and method of PSO-BPNN predicting model for activated carbon EDLC electrochemical properties were as follows.

6.1 BPNN Predicting Model

In order to predict activated carbon EDLC electrochemical properties, a mathematical model expressed electrochemical properties in terms of experimental data was required. In this study, the mathematical model was built using BP neural network. During the BPNN model design process, we focused on the following aspects:

a. Experimental Database. The first step of BPNN modeling was to build an input-output training database according to activated carbon EDLC electrode experimental data. The experiment process was detailed in section 2 of this paper. The experimental database was used to train and test BPNN model.

b. Input-output Vector. In order to identify input vector, influence factors about activated carbon EDLC electrochemical properties should be carefully analyzed. There are lots of influent factors and their mutual influences are even strong. How-

ever, among of them, activation dosage, activation time and activation temperature are three key factors, so they were all selected as input vectors. Output vectors are BET specific surface area(S_{BET}) and specific capacitance.

The first 18 experimental data (No.A_1 to No.A_{18} in Table 1) were used to train BPNN model, while the last 5 data (No.B_1 to No.B_5 in Table 1) were used to test BPNN model. In order to eliminate dimension different, all input and output data were normalized with the Eq.(10):

$$X_{norm} = \frac{2(X - X_{\min})}{X_{\max} - X_{\min}} - 1. \tag{10}$$

All input and output data are mapped to the range of [-1,1].

c.Transfer Function Selection. How to select transfer function and activation function of BPNN is important. There are many transfer functions for neural networks such as logarithmic, tangent hyperbolic and gauss. In this paper, among these transfer functions, tangent hyperbolic sigmoid (tansig) led to the minimum mean square error (MSE) and was selected as hidden layer transfer function therefore. Because log-sigmoid(logsig) function mapped neurons value well to [0,1], and it was selected as output layer transfer function .

Tansig function was represented as follow:

$$f(x) = 2/(1 + e^{-2x}) - 1. \tag{11}$$

Logsig function was represented as follow:

$$f(x) = 2/(1 + e^{-2x}). \tag{12}$$

d. Number of Hidden Layer Neurons. NN training seems to be highly sensitive to number of hidden layer neurons; however, the number of hidden layer neurons is usually determined by trial and error. It is known that too few neurons can lead to under-fitting whereas too many neurons can contribute to over-fitting. In this work, gradually growth method was used to train network until the number of hidden layer nodes met the requirements of error. During training process, number of hidden layer neurons had been increased from 5 to 9 in order to define output accurately. Statistical methods were used to compare the training results. Mean-squares error (MSE) was defined as follows:

$$MSE = \frac{1}{mp} \sum_{p=1}^{p} \sum_{s=1}^{m} (d_{ps} - y_{ps})^2, \tag{13}$$

where m is the number of output nodes, p is the number of training sample, $d_{ps}$is desired output value of NN model and $y_{ps}$is actual output value of NN model.

Figure 3 illustrated the performance of BPNN training with different numbers of hidden layer neurons. As seen from Figure 3, when the number of hidden layer neurons was 6, it met the requirements of model design because MSE of the NN model was close to 0.0012 and gradually fell to 0.001 after 150 epoch. Hence 6 were set as optimal number of hidden layer neurons.

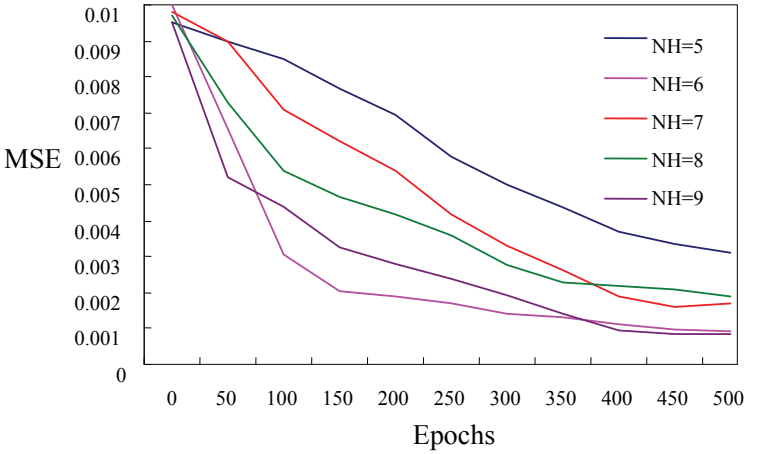

Figure 3: Relationship diagram among MSE, number of hidden layer neurons and iteration epochs

6.2 Optimized PSO-BPNN Predicting Modeling

BPNN structure was composed of three layers: three input layer neurons corresponding to activation dosage, activation time and activation temperature, two output layer neurons corresponding to BET specific surface area and specific capacitance and one hidden layer with 5-9 neurons. Table 2 summarized learning factors of BPNN model during training and testing processes.

Table 2: Learning factors of BPNN

Parameter item	Value
Learning rate (η)	0.04
Momentum constant	0.9
Maximum number of epochs	500
Goal of MSE	0.01

In PSO-BPNN model, all particles were encoded by real number strings. Each real number string represented weights of BPNN model. The improved PSO–BP predicting and optimizing modeling approach of activated carbon EDLCs' electrochemical properties can be described as follows:

Step 1. Set parameters for improved PSO.

This step included setting the numbers of PSO particles, dimension D, randomly initial string with adaptive group-scale, the initial w, learning factor c_1, c_2 and iteration stop conditions. In this study, s was set as 18, D was set as 47, w was set as 0.6, c_1 and c_2 were set as 2 and largest iterative number was set as 500.

Step 2. Calculate the fitness function value according to Eq.(14)

$$J = \frac{1}{2}\sum_{n}\sum_{k}(Y_{n,k} - \bar{Y}_{n,k}) \tag{14}$$

If the function value met certain conditions, training process would be terminated and we obtained output results. Otherwise the fitness information will be returned to the next step.

Step 3. Update ω according to fitness function.

Step 4. Return to the second step and calculate the new fitness with renewed ω.

*Step 5.*Get optimal solution.

When iteration was terminated, an improved PSO-BP algorithm was obtained based on initial optimal solution of improved PSO algorithm.

Step 6. The optimized PSO-BPNN model was used to predict activated carbon EDLCs' electrochemical properties.

6.3 PSO-BPNN Testing

The performance of trained PSO-BPNN model can be evaluated by errors of train and test sets. The PSO-BPNN model was tested by eighteen input data and five output data. For each data, the predicted values of electrochemical properties were compared with mean error percentage (MRE) and root mean squared (RMS). MRE and RMS were defined as follows respectively:

$$RMS = ((1/p)\Sigma|V_e - V_p|^2)^{1/2}, \tag{15}$$

$$MRE = (1/p)\,|(V_e - V_p)/V_e| \times 100\%, \tag{16}$$

where p is the number of experimental data, V_e is the experimental value of electrochemical properties, and V_p is the predicted value of electrochemical properties.

7 Results and Discussion

Comparison of experimental data and prediction data of train set were shown in Figure 4 when training MSE was 0.001. It showed that improved PSO-BPNN model were characterized by high training precision and well fitting prediction result (experimental data represented by "·" and prediction data represented by " ").

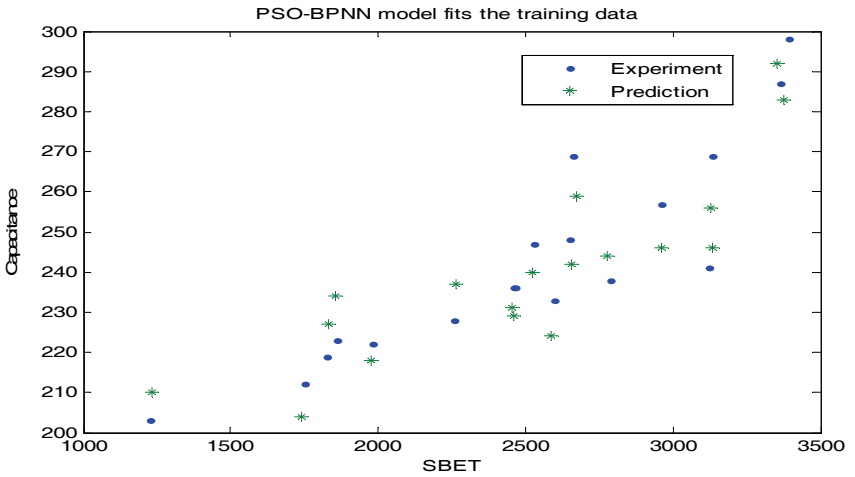

Figure 4: Comparison of experimental and predicted data of train set

Generalization ability is used to verify if the prediction data of test set and experimental data fit very well. The generalization ability of the system is more important. By comparing the prediction data with experimental data of test set, it could be clearly seen that experimental data were very close to the predicted data of test set from Figures 5 and 6.

RMS and MRE of train and test sets were shown in Table 3. It can also be realized that the PSO-BPNN model characterize perfect prediction and generalization performances.

Table 3: RMS and MRE of train and test sets

	RMS-train	MRE-train	RMS-test	MRE-test
S_{BET}	0.0018	0.0021	0.052	0.0102
Specific capacitance	0.0024	0.0023	0.065	0.0322

Before this study, activated carbon EDLCs' electrochemical properties prediction

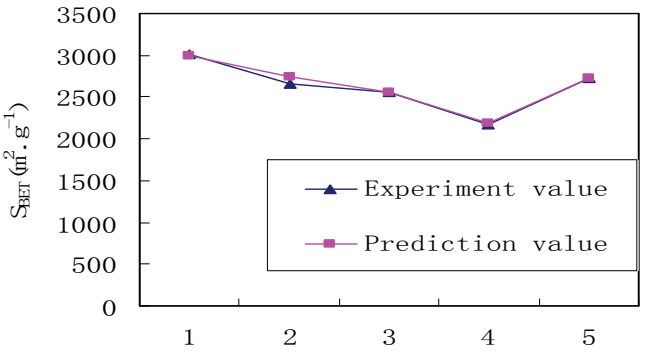

Figure 5: Comparison of experimental and predicted data of test set for S_{BET}

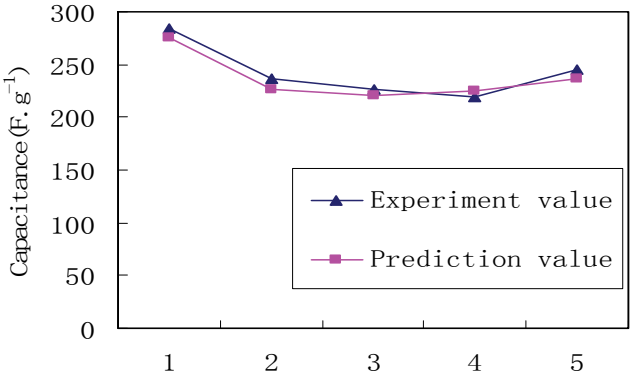

Figure 6: Comparison of experimental and predicted date of test set for specific capacitance

model based on BPNN had been established. To further verify the accuracy of the PSO-BPNN model, we compared the prediction results of BPNN model with the results of PSO-BPNN model. The comparison results were shown in Table 4.

It can be seen from the Table 4 that BPNN model did enough to meet a requirement for predicting application of activated carbon EDLCs' electrochemical properties,but the largest relative error percentage of BPNN model is more 5.5% than that of improved PSO-BPNN model and the average relative error percentage is more 5% than that of improved PSO-BPNN model. In the light of the maximum, minimum, RMS or average errors, the PSO-BPNN's are all less than BPNN's.In addition, during training and testing process, execution time of PSO-BPNN is quicker 3.4 times than BPNN's. It is clear that the prediction and generalization perfor-

Table 4: Comparison results of BPNN and improved PSO-BPNN

NO.	Experimental data		BP NN model				Advanced PSO-BP NN model			
			Predicted value		MRE		Predicted value		MRE	
	S_{BET}	Specific capacitance	S_{BET}	Specific capacitance	S_{BET}	Specific capacitance	S_{BET}	Specific capacitance	S_{BET}	Specific capacitance
B1	3013	284	2985	271	0.0113	0.0458	2993	276	0.0066	0.0282
B2	2663	237	2631	224	0.0105	0.0549	2750	226	0.0327	0.0464
B3	2559	226	2514	233	0.0176	0.031	2562	220	0.0012	0.0265
B4	2169	219	2126	210	0.0106	0.0411	2181	225	0.0055	0.0274
B5	2732	245	2704	235	0.0066	0.0408	2719	237	0.0048	0.0327
RMS			SBET 0.087		Specific capacitance 0.093		SBET 0.052		Specific capacitance 0.065	

mance of PSO BPNN model have been greatly improved.

8 Conclusions

The aim of this paper was to show material chemometrics and NNs intelligent theory application possibility for the activated carbon EDLCs' electrochemical properties. Results show that the NNs can be used as an alternative theoretic method in material systems. In this work, the electrochemical properties prediction model was developed and optimized using improved PSO BPNN algorithm. Based on experimentation and investigation carried out, the following conclusions are drawn:

a. NN model with three layers and one hidden layer of 6 neurons is a powerful method for predicting activated carbon EDLCs' electrochemical properties.

b. The combined improved PSO-BPNN model can be well used to optimize prediction model. The prediction model is characterized by good approximation performance as RMS-test of S_{BET} and RMS–test of specific capacitance are 0.052 and 0.065 respectively. MRE of S_{BET} and specific capacitance between experimental data and the predicted output are 1.02% and 3.22% respectively. Hence, prediction model for activated carbon EDLCs' electrochemical properties based on improved PSO-BPNN is possessed with good approximation, rapidly converging and high precision.

c. The advanced PSO-BPNN modeling method is simple, rapid and reliable. The essential principle can be popularized to prediction and simulation of other material system.

In addition, by defining more impact factors and more experimental data, the improved PSO-BPNN model would present better prediction and generalization performance, however, this would increase experiment cost and time.

Although PSO-BPNN is an efficient intelligent theory method for activated carbon EDLCs' electrochemical properties, it should consider more variables of experimental conditions. These extensions will be the focus of our future work.

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