J. LI, C. F. ZHANG, R. M. YIN, W. H. ZHANG

ISSN 0543-5846
METABK 57(4) 265-268 (2018)
UDC – UDK 666.5–661.68.86.931.938/.001.1:536.45:620.17:531.75=111

PREDICTION OF MECHANICAL PROPERTIES OF β-SiAlON CERAMICS BASED ON BP NEURAL NETWORK

Received – Primljeno: 2018-03-18
Accepted – Prihvaćeno: 2018-06-10
Original Scientific Paper – Izvorni znanstveni rad

β-$Si_{6-Z}Al_ZO_ZN_{8-Z}$ (0<Z≤2) ceramics with rod-like grain morphology were prepared by gas pressure sintering, and their mechanical properties (i.e., bulk density, hardness, fracture toughness, and flexural strength) were evaluated. A model to predict the mechanical properties of β-SiAlON ceramics was established by back propagation (BP) neural network, and the relationships between process parameters (i.e., Z-value and temperature) and mechanical properties were investigated. Results show that the model had good prediction accuracy and maximum relative error lower than 8 %. The model could reflect the complex nonlinear relationship between the process parameters and the mechanical properties of β-SiAlON ceramics. The model can provide an effective reference for optimizing the design of β-SiAlON ceramics.

Keywords: β-SiAlON ceramics, BP neural network, temperature, mechanical property, density

INTRODUCTION

β-SiAlON is a solid solution of β-Si_3N_4, where Si and N are partially replaced by Al and O and its general formula is $Si_{6-Z}Al_ZO_ZN_{8-Z}$ (0<Z<4,2). β-SiAlON has a hexagonal crystal structure with rod-like grain morphology, resulting in very good toughness and strength [1]. Moreover, β-SiAlON has the best oxidation resistance, high-temperature resistance, and thermal stability among SiAlON ceramic series [2]. In recent years, β-SiAlON and β-SiAlON composite ceramics have been widely applied in high-temperature metallurgy, materials, aerospace, military, and other related fields because of their excellent physicochemical properties [3].

Back propagation (BP) neural network, which is one of the most extensively used artificial neural networks, has good associative memory and high tolerance. Moreover, this process can quickly and effectively reflect highly nonlinear and complex relationships between influencing factors. BP neural network has been preliminarily applied in SiAlON ceramic materials. Zhang et al. [4] used this network to predict the relative content of α-phase of Ca-α-SiAlON prepared by carbothermal reduction and nitridation synthesis under different process conditions. Li et al. [5] introduced the BP neural network into the design of β-SiAlON ceramic prepared by microwave-assisted carbothermal reduction and predicted the relative content of the β-phase under various ratios of n(Si)/n(C) and temperatures.

In this study, β-SiAlON ceramics with rod-like grain morphology were prepared by gas pressure sintering using α-Si_3N_4, Al_2O_3 and AlN as raw materials. A model to predict the mechanical properties of β-SiAlON ceramic materials was established using the BP neural network. The relationships between process parameters (i.e., Z-value (0<Z≤2) and temperature) and mechanical properties (density, hardness, fracture toughness, and bending strength) were investigated.

EXPERIMENTAL PROCEDURE

Material preparation

The starting composition was prepared using α-Si_3N_4 (UBE-10, containing 1,6 % oxygen), Al_2O_3 (99,99 %; Sumitomo AES IIC), and AlN (Tokuyama, containing 1 % oxygen). In addition, 6 wt.% Y_2O_3 was added to promote the sintering densification. Starting compositions of all samples are given in Table 1.

An α-Si_3N_4 suspension with concentration of 28,8 g/L with absolute ethanol as a solvent was prepared. Then, Al_2O_3, AlN, and Y_2O_3 were added into the suspension. A 5-mL volume of PEG400 was used as a dispersant. The slurry was agitated, ultrasonically dispersed for 60 min, and subsequently dried at 500 °C. After drying, the composite powders were milled and sieved to 100–150 mesh, pressed uniaxially with maximum pressure of 25 MPa, and subsequently cold isostatically pressed at 300 MPa to form dense green compacts. Then, the green compact was sintered by gas pressure sintering. The heating rate was 10 °C/min. The final sintering temperature (T) was 1 550–1 800 °C and holding time was 2 h. Nitrogen (2 MPa) was introduced into the furnace at 1 150 °C, and the cooling rate was set to 15 °C/min.

J. Li (133501020@csu.edu.cn), C. F. Zhang, W. H. Zhang, School of Metallurgy and Environment, Central South University, Changsha, China. R. M. Yin, College of Metallurgical Engineering, Hunan University of Technology, Zhuzhou, China

Table 1 **Composition and denomination of samples**

Z value	Composition / wt. %		
	Si_3N_4	Al_2O_3	AlN
Z=0,3	69,09	2,55	1,04
Z=0,6	66,61	4,24	1,74
Z=1,0	60,45	8,47	3,47
Z=2,0	48,19	16,88	6,92

Characterization techniques

The phase composition was identified by X-ray diffraction (XRD; Rigaku D/MAX-IIIC, Japan) with Cu radiation at 40 kV and 50 mA at a scan rate of 4 °/min to record the diffraction patterns. The microstructures of the polished surface were observed by scanning electron microscopy (SEM; SIRION 200, FEI, USA). The bulk density (ρ) was measured according to Archimedes' principle in deionized water. Room-temperature Vickers hardness (HV) was tested by a Vickers diamond indenter (HV-50). The standard procedure involved the application of a load of 10 kg for 15 s. The averages of the single-edge notched beam (SENB) were used to measure room-temperature fracture toughness (K_{Ic}) in a universal material testing machine (Instron-5569). The flexural strength (σ) was measured by three-point bending method on an electronic universal testing machine (CSS-44100).

Table 2 **Experimental results of the mechanical pro- perties of the samples**

Sample	Z	T / °C	ρ / kg·m^{-3}	HV / GPa	K_{Ic} / MPa·m$^{1/2}$	σ / MPa
1#	0,3	1 550	3,090	1 326	6,821	530
2#	0,3	1 600	3,090	1 389	6,878	715
3#	0,3	1 650	3,210	1 515	6,934	871
4#	0,3	1 700	3,239	1 582	7,662	591
5#	0,3	1 750	3,246	1 593	6,916	507
6#	0,3	1 800	3,214	1 507	7,676	670
7#	0,6	1 550	3,240	1 634	7,375	501
8#	0,6	1 600	3,240	1 600	7,565	806
9#	0,6	1 650	3,240	1 589	7,580	710
10#	0,6	1 700	3,225	1 571	7,876	704
11#	0,6	1 750	3,229	1 627	7,245	901
12#	0,6	1 800	3,228	1 549	6,997	659
13#	1,0	1 550	3,210	1 546	6,237	550
14#	1,0	1 600	3,210	1 538	6,246	780
15#	1,0	1 650	3,210	1 528	6,558	740
16#	1,0	1 700	3,205	1 525	6,564	732
17#	1,0	1 750	3,206	1 514	6,668	729
18#	1,0	1 800	3,150	1 507	6,798	666
19#	2,0	1 550	3,150	1 421	4,370	414
20#	2,0	1 600	3,150	1 442	5,403	463
21#	2,0	1 650	3,150	1 543	5,730	436
22#	2,0	1 700	3,133	1 458	5,720	363
23#	2,0	1 750	3,170	1 447	5,640	346
24#	2,0	1 800	3,089	1 429	6,111	234

XRD results showed that all samples were single-phase β-SiAlON (β-$Si_{5,7}Al_{0,3}O_{0,3}N_{7,7}$ when Z=0,3, β-$Si_{5,4}Al_{0,6}O_{0,6}N_{7,4}$ when Z=0,6, β-Si_5AlON_7 when Z=1,0, β-$Si_4Al_2O_2N_6$ Z=2,0). The SEM results showed that all samples had rod-like microstructure morphology. The test results of bulk density, Vickers hardness, fracture toughness, and bending strength of all samples are listed in Table 2. Figure 1 shows the typical microstructures of several samples under different Z values and sintering temperatures. The grain size distribution varied from 600 nm to 20 μm.

BP NEURAL NETWORKS

BP neural network is a feed-forward neural network with three or more layers. Each layer consists of several neurons (Figure 2).

The steps of the BP neural network are as follows:

(1) Sample standardization

The input sample is defined as $X=(X_1, X_2, \ldots, X_n)$, and the corresponding expected output is $Y=(Y_1, Y_2, \ldots, Y_n)$. The raw data should be normalized according to Equation (1) to obtain the ideal training effect.

$$x_i^{new} = \frac{x_i^{old} - \min\left(x_i^{old}\right)}{\max\left(x_i^{old}\right) - \min\left(x_i^{old}\right)}, \quad i = 1 - n \qquad (1)$$

Where x_i^{old} is the raw data, x_i^{new} is the dimensionless data, and n is the number of experimental data.

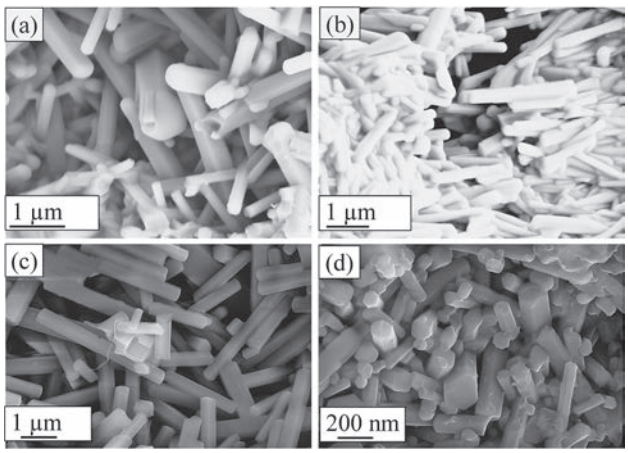

Figure 1 Typical microstructure (a) 4#, (b) 12#, (c) 17#, (d) 21#

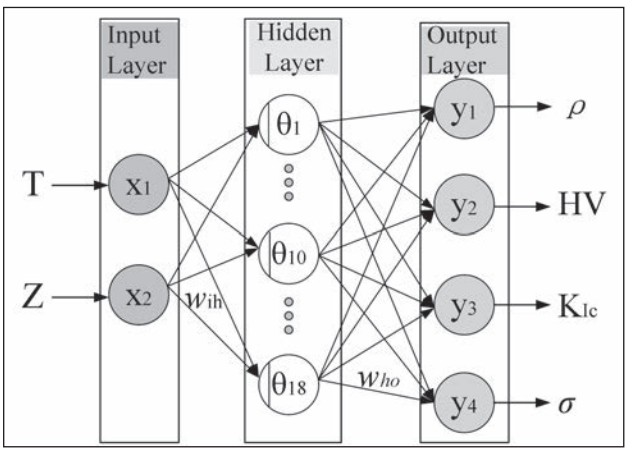

Figure 2 Schematic of BP neural network

(2) Initialization of the weight coefficient

A small non-zero random number is assigned to the connection weight (w_{ij}) of each layer, and a random value of (−1, 1) interval is assigned to the threshold.

(3) Calculation of the output of each layer

The network output values are calculated by the excitation function known as Sigmoid function. The input and output of each node of the hidden layer (k<m) are defined as follows:

$$s_i^k = \sum_{j=1}^{p} x_j^k w_{ij}^k - \theta_i, \quad b_i^k = \frac{1}{1+e^{-s_i^k}} \tag{2}$$

The input and output of each node of the output layer (k=m) are described by Equation (3) as follows:

$$l_i^k = \sum_{j=1}^{q} b_i^k w_{ij}^k, \quad c_i^k = \frac{1}{1+e^{-l_i^k}} \tag{3}$$

Where m is the number of layers; p and q are the numbers of neurons in the input and hidden layers, respectively; s_i^k, l_i^k are the inputs of the i-th neuron in the k-th layer; b_i^k, c_i^k are the outputs of the i-th neuron in the k-th layer; w_{ij}^k is the connection weight; and θ_i is the threshold of a nerve cell.

(4) Calculation of the learning error

$$E_k = \sum_{i=1}^{q} \left(y_i^k - c_i^k \right)^2 / 2 \tag{4}$$

Where y_i^k is the desired output of the network, and E_k is the mean square of the deviation between the network's expected output and the calculated output.

The learning error for each node of the output layer (k=m) is as follows:

$$\delta_i^m = \left(y_i^m - c_i^m \right) c_i^m \left(1 - c_i^m \right) \tag{5}$$

The learning error for each node of the hidden layer (k<m) is calculated as follows:

$$\delta_i^k = b_i^k \left(1 - b_i^k \right) \sum_{j=1}^{q} w_{ij}^{k+1} \delta_j^{k+1} \tag{6}$$

(5) Correct weight coefficient and threshold

The momentum method is used to speed up the convergence and prevent oscillation.

$$w_{ij}^k (t+1) = -\eta \delta_i^k c_j^{k-1} + \alpha \Delta w_{ij}^k (t) \tag{7}$$

Where η is the learning rate, α is the momentum coefficient, and t is the iteration times.

When the network is trained, the errors of the neurons in the output layer propagate back to the hidden and input layers, and the weights between the layers are constantly corrected. If the requirements are satisfied, the algorithm ends. Otherwise, step 3 is repeated until the system's mean square error is minimized. Using the resulting weights after training, a model can be constructed to predict the relationship between sintering process and mechanical properties of β-SiAlON.

RESULTS AND DISCUSSION

From the 24 sets of experimental data, 20 groups were randomly selected as learning samples, and the remaining 4 groups were the verification groups. A three-layer (i.e., input, intermediate, and output layers) BP neural network with two input parameters (Z value and temperature) and four output parameters (bulk density, Vickers hardness, fracture toughness, and flexural strength) were designed and tested repeatedly. Finally, a single hidden layer structure of 18 neurons was selected.

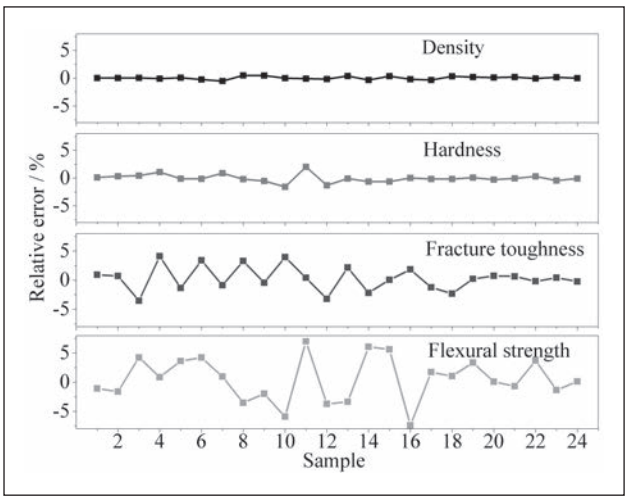

Figure 3 Variation in the relative error with testing data

Figure 3 shows the variation in the relative errors with the testing data. The experimental results of density, Vickers hardness, and fracture toughness are in good agreement with the predicted values of the model, with maximum relative error of less than 5 %. However, the relative error between the predicted value and experimental results of bending strength is relatively larger, but the maximum relative error is also less than 8 %. This finding indicates that the established model has good fitting precision. Therefore, the proposed model can be used to effectively predict and investigate the nonlinear relationship between the process parameters and the mechanical properties. The model can be used as a reference to optimize the design of β-SiAlON ceramics.

The relationship between the process parameters and the mechanical properties, namely, density, Vickers hardness, fracture toughness, and flexural strength, of the β-SiAlON ceramics are shown in Figures 4–7, respectively. Increasing the Z value resulted in the initial increase and subsequent decrease in the bulk density, hardness, fracture toughness, and bending strength. However, no obvious linear relationship is observed between temperature and the mechanical properties. When the Z value is in the range of 0,45–1,0 and the sintering temperature is 1 600–1 750 °C, the β-SiAlON ceramics can obtain relatively high bulk density, hardness, fracture toughness, and bending strength with the highest value of 3,26 kg·m^{-3}, 1 630 GPa, 7,78 MPa·m$^{1/2}$, and 914 Mpa, respectively. Therefore, β-SiAlON ceramics prepared using gas pressure sintering within this process range are most likely to yield the best mechanical properties. Using this model, the Z value and sintering temperature can be well determined according to the re-

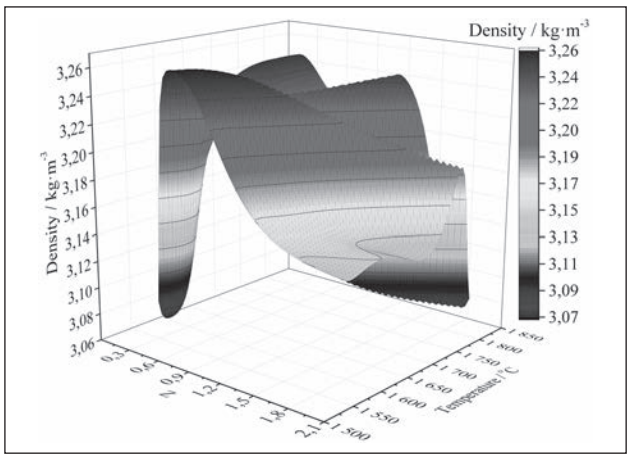

Figure 4 Bulk density versus Z value and temperature

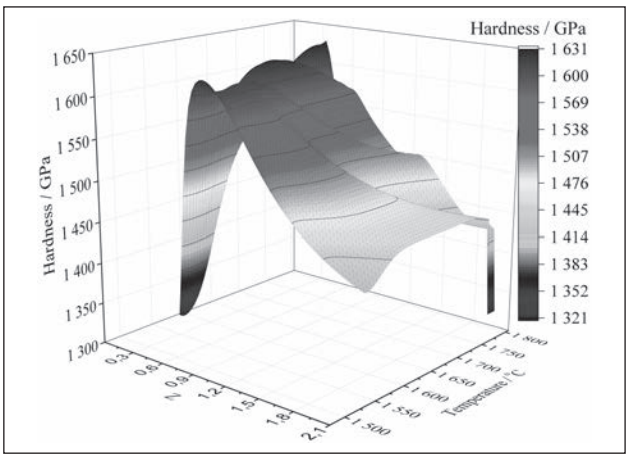

Figure 5 Hardness versus Z value and temperature

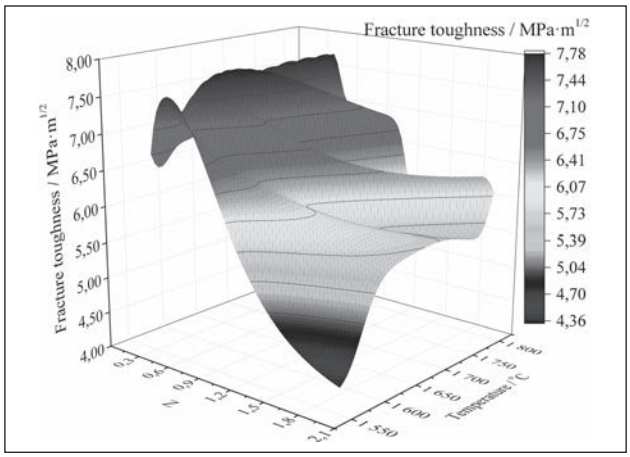

Figure 6 Fracture toughness versus Z and temperature

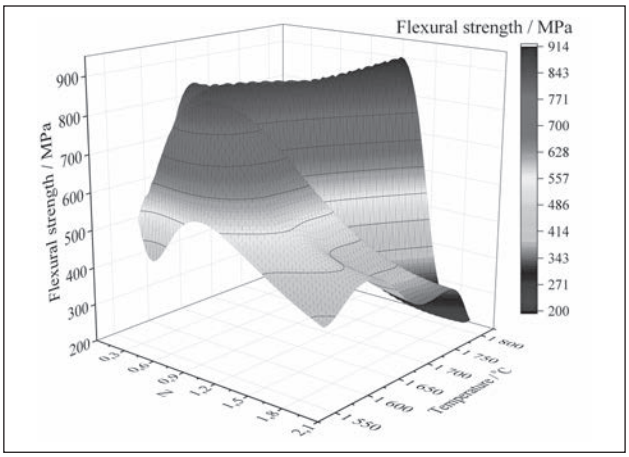

Figure 7 Flexural strength versus Z and temperature

quired performance, greatly reducing the amount of experiments and saving costs.

CONCLUSIONS

In this study, β-SiAlON ($0<Z\leq2$) ceramics with rod-like grain morphology were prepared by gas pressure sintering. A model to predict the mechanical properties of β-SiAlON ceramics was established using BP neural network method. The relationships between process parameters (i.e., Z value and temperature) and mechanical properties (i.e., density, hardness, fracture toughness, and bending strength) were investigated. The following conclusions are drawn:

(1) The model has good prediction accuracy, with maximum relative error of less than 8 %. This result indicates that the model can effectively describe the complex nonlinear relationship between the Z value and temperature versus the bulk density, hardness, fracture toughness, and bending strength of the β-SiAlON ceramics.

(2) When the Z value is in the range of 0,45–1,0 and sintering temperature is 1 600– 1 750 °C, the β-SiAlON ceramics can obtain relatively high bulk density, hardness, fracture toughness, and bending strength with highest values of 3,26 $kg \cdot m^{-3}$, 1 630 GPa, 7,78 $MPa \cdot m^{1/2}$, and 914 Mpa, respectively.

Acknowledgements

This work is financially supported by the National Natural Science Foundation of China (No. 51574120).

REFERENCES

[1] N. Calis Acikbas, O. Demir, The effect of cation type, intergranular phase amount and cation mole ratios on z value and intergranular phase crystallization of SiAlON ceramics, Ceramics International 39(2013), 3249-3259.

[2] X. Yi, W. Zhang, T. Akiyama, Thermal conductivity of β-SiAlONs prepared by a combination of combustion synthesis and spark plasma sintering, Thermochimica Acta 576(2014), 56-59.

[3] X. Hou, X. Lu, B. Peng, B. Zhao, M. Zhang, K. Chou, A new approach to interpreting the parabolic and non-parabolic oxidation behaviour of hot-pressed β-SiAlON ceramics, Corrosion Science 58(2012), 278-283.

[4] H. Zhang, S. Du, Y. Cao, L. Lu, S. Zhang, Investigation of preparation of Ca-α-Sialon powders via artificial neural networks analysis, Ceramics International 40(2014), 2287-2293.

[5] F. Li, F. Fu, L. Lu, H. Zhang, S. Zhang, Preparation and artificial neural networks analysis of ultrafine β-Sialon powders by microwave-assisted carbothermal reduction nitridation of sol–gel derived powder precursors, Advanced Powder Technology 26(2015), 1417-1422.

Note: The professional translator for the English language is Jing Li, Changsha, China.