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Artificial neural network modeling of ceramics powder preparation: Application to $NiNb_2O_6$

W. Laosiritaworn [a,*], O. Khamman [b], S. Ananta [b], R. Yimnirun [b], Y. Laosiritaworn [b]

[a] Department of Industrial Engineering, Faculty of Engineering, Chiang Mai University, Chiang Mai 50200, Thailand
[b] Department of Physics, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand

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Abstract

This paper studied the modeling of the synthesis process of $NiNb_2O_6$ (NN) powder using an artificial neural network (ANN). The characteristic of interest was the amount of NN phase percentage produced from the synthesis process. Three controlling factors affecting the mentioned characteristic were dwell time, calcined temperature and heating/cooling rate. Design of experiments (DoE) technique was used to analyze the relationship of controlling factors to the amount of NN phase. The results show that calcined temperature is the most important factor affecting the amount of NN phase. The dwell time and heating/cooling rate are less significant on the phase but longer dwell time and higher heating/cooling rate are appreciable for the slightly higher purity. Multiple regression was also used to compare the results and the ANN was found to significantly outperform the regression analysis.

Keywords: Powders preparation; Artificial neural network

1. Introduction

Typically, in preparing ceramic powder, several techniques ranging from mechanical milling to chemical method can be used [1]. However, due to many factors involved such as milling time, dwell time, calcined temperature, heating/cooling rate, etc., lots of experimental combinations are possible. Therefore, to search for the optimal condition in obtaining the powder with a pure preferred phase at low cost could be difficult. This method of trial-and-error experiment is not economical and the optimal experimental setting cannot be guaranteed. Consequently, there is a need for a technique which can be used to find the relation between the relevant parameters to obtain the powder with a high purity phase in a cost efficient way. Thus, there comes the objective of this study to suit such a situation using an artificial neural network.

Artificial neural network (ANN) is one of several artificial intelligence tools which has been widely used in various applications due to its ability to learn from samples, and fault tolerance. In this study, it is used to predict the percentage of the desired material in ceramic powder preparation. The $NiNb_2O_6$ (NN) powder was chosen as a case study due to its interesting in being used as a precursor to prepare $Pb(Ni_{1/3}Nb_{2/3})O_3$ [9] which is a potential candidate for electroceramics applications [1–3].

In this study, by following the NN powder preparation technique suggested in Ref. [9], the percentage of the obtained NN phase relevant to the condition of preparation including dwell time, calcined temperature and heating/cooling rate was investigated. Fifty-one experimental samples have been used to construct the ANN model. Once the ANN has been assembled, it was used to predict the percentage of NN phase of the unseen input conditions. Therefore, this minimizes the need to conduct an actual experimentation. Design of experiments (DoE) was then used to analyze the effect of controlling factors to the percentage of NN phase. Multiple regression model was also used for a comparison purpose.

1.1. Background theories

1.1.1. Artificial neural network

Artificial neural network (ANN) is ‘an interconnected assembly of simple processing elements, *units* or *nodes*, whose

* Corresponding author.
E-mail address: wimalin@eng.cmu.ac.th (W. Laosiritaworn).

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functionality is loosely based on the animal neuron. The processing ability of the network is stored in the inter-unit connection strengths, or *weights*, obtained by a process of adaptation to, or *learning* from, a set of training patterns' [4].

Inspired by how the brain and nerve cells work, ANN is constructed by connecting simple processing elements or neurons together. Neurons can be located in the input layer, the output layer or the hidden layers. The input layer takes data from the outside of a neural network and sends it to hidden layer neurons. The output layer sends the information back outside the network.

ANN training can be supervised or unsupervised (self-organising). This paper is focused on supervised training. The back-propagation algorithm is the most extensively adopted algorithm for network training [5] and is the type of network used in this research.

1.1.2. Statistical design of experiments

Statistical DoE was first developed in the early 1920s by Sir Ronald Fisher to determine the effect of multiple factors on the outcome of agricultural trials. The work of statisticians [6,7] with an interest in this area has provided a firm foundation for practitioners. DoE is a statistical and structural method used to analyze relationships between factors affecting processes (independent variables) and the output of those processes (dependent variables). DoE techniques are widely used, both by researchers, for characterisation, optimisation, and modelling [8].

2. Experimental procedures

2.1. NN powder preparation

The NN powder was synthesized by the solid-state reaction of thoroughly ground mixtures of NiO and Nb_2O_5 powders milling in the required stoichiometric ratio. The detailed description of the NN powder preparation is given elsewhere [9]. In this study, the NN powder was synthesized by varying dwell time from 1 to 48 h, calcined temperature from 500 to 1200 °C, and heating/cooling rate from 3 to 30 °C/min. Then the obtained powder is investigated by X-ray diffraction (XRD) technique to calculate the percentage of NN phase. However, if there exist more than two phases in the XRD pattern, the NN percentage is set as unclassified. Controlling factors in the experiments were set by trial-and-error and percentage of NN phase were recorded. Fifty-one records of experimental data were available.

2.2. Multiple regression analysis

A multiple regression model has the following form:

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \cdots + \beta_k x_k,$$

where β_0 is the intercept, $\beta_1, \ldots, \beta_k$ refer to parameters representing the contribution of the independent variables, and $x_1, \ldots, x_k$ are independent variables.

Of the 51 records available, 41 records were randomly selected and used to fit regression equation while the remaining 10 records was used as independent test set. The regression equation is as follows:

$$y = -103.905 + 0.189x_1 + 0.810x_2 + 0.616x_3,$$

where y is the percentage of NN phase obtained, while x_1, x_2 and x_3 refer to calcined temperature, dwell time and heating/cooling rate, respectively. The results from regression analysis are compared with the result from ANN in the next section.

2.3. Artificial neural network training and testing

The same data set used to fit regression equation was used to train ANN. The training was carried out with 41 records and the remaining 10 records for testing ANN performance. Qnet v2000 software was used to develop an ANN model. The network developed consisted of three layers. Input layer has three neurons representing calcined temperature, dwell time and heating/cooling rate while the output layer has one neuron which represents the prediction of percentage of NN phase obtained. Hidden layer neurons were selected by trial-and-error and the appropriate number of hidden neurons were 17. Root mean square error of the training and testing data were 0.039701 and 0.049907, respectively, and the correlation coefficient of training and testing data were 0.977587 and

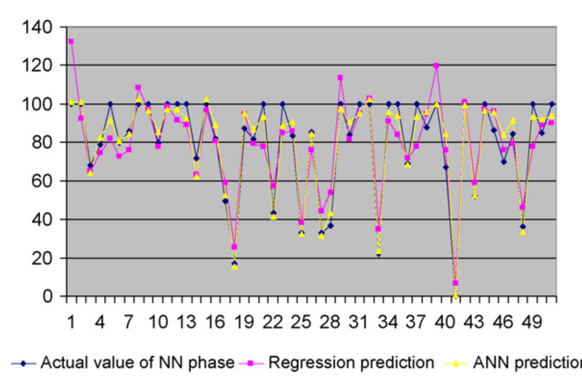

Fig. 1. The comparison of actual value with regression prediction and ANN prediction.

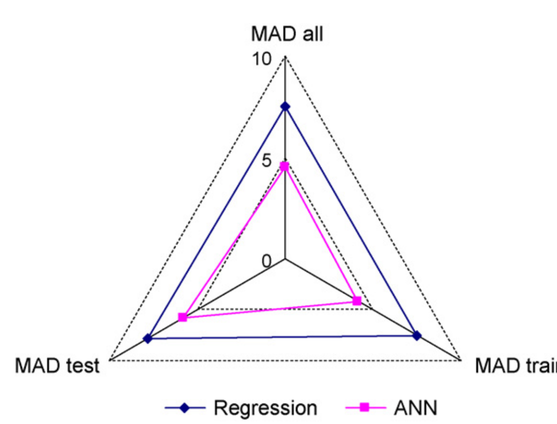

Fig. 2. MAD of results from regression model and ANN model.

Table 1
ANOVA analysis

Source	d.f.	Seq SS	Adj SS	Adj MS	F	P
Calcined temperature	4	148162.3	148162.3	37040.6	3532.83	0
Dwell time	6	2207	2207	367.8	35.08	0
Heating/cooling rate	2	525.1	525.1	262.6	25.04	0
Calcined temperature × dwell time	24	280.9	280.9	11.7	1.12	0.363
Calcined temperature × heating/cooling rate	8	2907.1	2907.1	363.4	34.66	0
Dwell time × heating/cooling rate	12	1317.7	1317.7	109.8	10.47	0
Error	48	503.3	503.3	10.5		
Total	104	155903.4				

d.f.: degree of freedom; Seq SS: sequencial sum of squares; Adj SS: adjusted sum of squares; Adj MS: adjusted mean square; F: F-statistic; P: p-value.

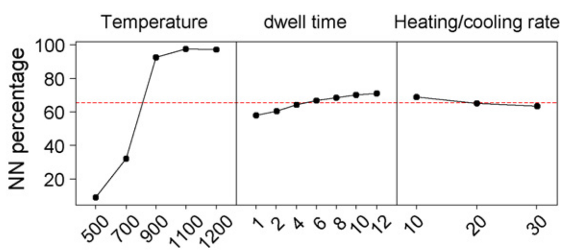

Fig. 3. Main effect plot (data means) of NN percentage.

0.947345, respectively. As the correlation coefficient of both training and testing data were very close to 1, the model is a good approximation of real experimentations.

Fig. 1 shows the actual data of percentage of NN phase compared with the prediction from regression and ANN. Fig. 2 is the radar chart that illustrates mean absolute deviation (MAD) of the overall, training and testing data of the results from both regression and ANN. It can be concluded from both figures that ANN significantly outperform multiple regression, as a result ANN model was used for further analysis.

2.4. Design of experimental analysis

After the ANN had been properly trained, it was used to predict experimental results. Experiments were set at five levels for the calcined temperature ranging from 500 to 1200 °C, seven levels for the dwell time ranging from 1 to 12 h and three levels for the heating/cooling rate ranging from 10 to 30 °C/min, with all possible combinations of experimental setting of 105. These experimental settings were fed to the trained ANN to obtain the prediction of the NN phase.

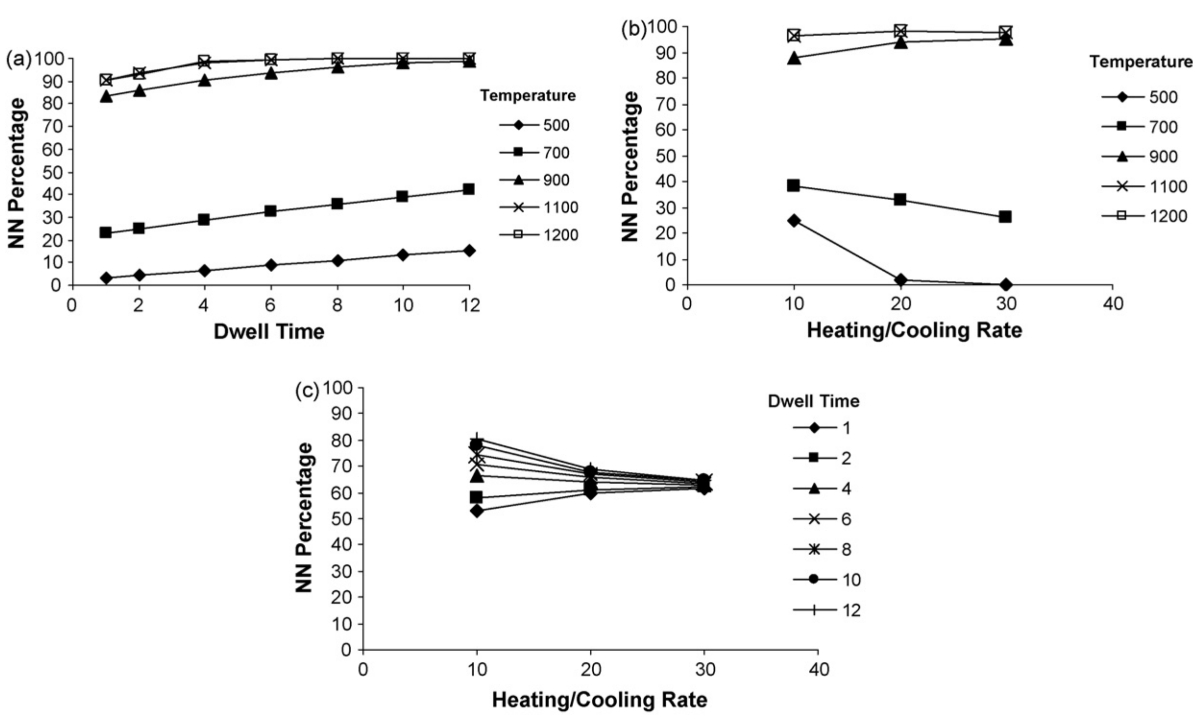

Fig. 4. Interaction plot (data means) of NN percentage: (a) between temperature and dwell time; (b) between temperature and heating/cooling rate; (c) between heating/cooling rate and dwell time.

3. Results and discussion

Analysis of variance (ANOVA) was carried out to identify significant factors and factor interactions to the percentage of NN. The ANOVA analysis was prepared by using MINITAB release 14. Table 1 summarises the significant factors of each response at the 95% confidence level. Significant factors at the 95% confidence level were the factor that has P value lower than 0.05. As a result, calcined temperature, dwell time, heating/cooling rate, interaction of calcined temperature and heating/cooling rate and interaction of dwell time and heating/cooling rate are found to be significant with different levels.

The relation among the experimental parameters in obtaining the high purity NN phase can be analysed by the main effect plot (Fig. 3), which shows the effect of each controlling factors on the NN percentage. The graph indicated that calcined temperatures has the highest effect on NN percentage and there is a steep decrease in NN percentage at temperature below 900 °C. Dwell time and heating/cooling rate have less effect than calcined temperature but their effects are still significant according to the ANOVA analysis.

The interaction plot (data mean) of NN percentage is shown in Fig. 4. Each sub-figure represents the interaction between a pair of preparation conditions in obtaining the NN phase. Since there are three relevant parameters considered in this study, when a pair is chosen, each data point in the considered sub-figure is calculated from the average over the remaining parameter. For instance, Fig. 4(a) shows the interaction of temperature and time of the obtained NN phase in which each data point is calculated from the average over all considered heating/cooling rate.

It can be summarised from Fig. 4(a) that the calcined temperature has prominent effect on the percentage of NN phase, where a very high percentage is obtained only for the temperature above (or equal to) 900 °C. A huge leap in the percentage is found in calcined temperature between 700 and 900 °C, but the longer dwell time only slightly enhances the percentage. Slight dependence on heating/cooling rate of the NN phase is also found (Fig. 4(b)). The strong reliance on the temperature is again apparent. On the other hand, Fig. 4(c) shows the relation of heating/cooling rate of the NN phase with varying dwell time. As can be seen, the high percentage is found for low heating/cooling rate but high dwell time. The description underlying all these phenomena can be given via thermodynamics of phase transformation. Presumably, to grow the NN phase, the thermal energy must be at the right condition corresponding to the energy barrier. If the calcined temperature is too low, the thermal energy might not be enough to trick out the NN phase. However, even the heating/cooling rate does not have a significant effect on the percentage of NN phase, but when the rate is too high, there might not be enough time for the NN phase to get growing so a longer dwell time is needed.

4. Conclusions

The ANN technique was used to study the relation among calcined temperature, dwell time and heating/cooling rate in obtaining the high percentage of NN phase. In this study, the high accuracy ANN model of NN synthesis process, was constructed from a minority of training examples, in predicting the results from an untrained experiment. Also from the prediction, a significant effect from the relevant parameters to the percentage of NN phase is apparent. The most important factor to obtain the high percentage is the calcined temperature while the dwell time has a moderate effect on the phase, and the heating/cooling rate has the least significant. The major advantage of ANN is that it allows the through analysis of the relationship of controlling factors to the amount of NN phase without conducting experiments randomly. The traditional trial-and-error experiment might results in conducting many experiments, yet the optimal solution might never be found. The techniques adopted in this article could also be applied to other process which will results in a more economical and structure way for data analysis.

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