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Application of neural network technique to high energy milling process for synthesizing ZnO nanopowders †

O. M. Lemine[1,*] and M. A. Louly[2]

[1]*Physics department, College of Sciences, Alimam Mohammad Ibn Saud Islamic University (IMSIU), Riyadh, Saudi Arabia*
[2]*Industrial Engineering Department, College of Engineering, King Saud University, Riyadh 11421, Saudi Arabia*

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Abstract

An artificial-neural-network (ANN) model was developed to estimate the crystalline size of ZnO nanopowder as a function on the milling parameters such as milling times and balls to powder ratio. This nanopowder was synthesized by high energy mechanical milling and the required data for training were collected from the experimental results. The synthesized ZnO nanoparticles are characterized by X-ray diffraction (XRD) and scanning electron microcopy (SEM). It was found that artificial neural network was very effective providing a perfect agreement between the outcomes of ANN modeling and experimental results with an error by far better than multiple linear regressions. An optimization model and this experimental validation of the ball milling process for producing the nanopowder ZnO are carried out.

Keywords: Mechanical milling; Nanoparticles; Neural network; Optimization; Simulation

1. Introduction

In general, nanocrystalline semiconductors with dimension less than 100 nm represent a relatively new class of materials. Their short-range structures are essentially the same as bulk semiconductors but their optical and electronic properties are dramatically different. In particular, they have been intensively studied because of the nano-effects, and quantum-like effects which can change several properties such as, photo catalysis, nonlinear optical properties and photoconductivity. It interesting also to notice that there are some recent researches show that ZnO nanomaterials has an applied aspects such as in solar energy conversion, photocatalysis, light-emitting materials [1], transparent UV protection films and chemical sensors [2], etc. Various techniques have been used to synthesize ZnO nanoparticles and can be categorized into either chemical or physical methods [3]. For example, hydrothermal [4], solvothermal [5], sol-gel [6], direct chemical synthesis [7] and ball milling [8], bubble electrospinning [22], etc. Among these synthetic routes, mechanical milling has proved to be an effective and simple technique to produce nanocrystalline powders and the possibility of obtaining large quantities of materials. However, properties of nanopowders obtained by milling method are affected by various parameters such as milling time, ball to powder mass ratio, rotation speed, balls diameters, etc. Several groups were interested by ball milling process modeling, mainly based on the mechanistic [9-13] and thermodynamic [14-16] approaches to achieve a general understanding at the atomic and phenomenological level. Recently, artificial neural network (ANN) becomes one of the most powerful modelling techniques in conjunction with the statistical approach. It is suitable for simulations of the correlations which are hard to be described by physical models [17]. The advantages of ANN modeling are reduction of time and cost in all the required experimental activities. Nguyen et al. [20] used artificial neural network for the prediction of deformations of steel plate. Ramakrishnan et al. [21] used similar approach for the optimization of operating parameters and performance evaluation of forced draft cooling tower. ANN can be used for the prediction of the mechanical milling outputs. However, there only limited work on the application of neural networks in the field of mechanical milling [18, 19]. Dashtbayazi et al. [18] have developed an artificial-neural-network model for modeling of mechanical alloying (MA) process. Their research mainly discussed the effects of ball milling parameters on the characteristics of the as-milled metallic powders. Some property predictions of the powders can be achieved through the model. Ma et al. [19] have studied the artificial-neural-network modeling of the mechanical alloying (MA) process. They used an artificial neural network model for modeling the effects of mechanical alloying pa-

*Corresponding author. Tel.: +96 6112586775, Fax.: +96 6112581614
E-mail address: leminej@yahoo.com
† Recommended by Associate Editor Dong Geun Lee

rameters including milling time, milling speed and ball to powder weight ratio on the characteristics of WC-18at.%MgO nanocomposite powders. Their model can be used for the prediction of properties of composite WC-MgO powders at various milling parameters.

The aim of this work is to develop a neural network model to the prediction and optimization of the ball milling process for synthesizing ZnO nanocrystalline. The process parameters, including milling times and ball-to-powder weight ratio are applied to the neural network inputs to provide information relating to particles size. The network is then trained to output the prediction on the powders particles size. An optimization model is then developed to find the best parameters producing the minimal average grain size.

2. Experimental

2.1 Samples preparation

Commercially obtained ZnO powders with average particle size of about 1 μm and 99.9% of purity, were introduced into a stainless steel vials with stainless steel balls (12 mm and 6 mm in diameter) in a SPEX 8000 mixer mill. The milling time and mass ratio of ball to powder were varied in the range of 1-5 h and 10-20, respectively.

2.2 Structural characterisation

X-ray powder diffraction (XRD) measurements were performed using Shimadzu diffractometer (θ-2θ) equipped with Cu-Kα radiation (λ = 1.5418 Å). It is known that X-ray diffraction line broadening is influenced by the particles size and the internal strains.

The crystalline size was calculated from the Schererrer formula:

$$D = \frac{K\lambda}{B\cos\theta}. \quad (1)$$

In this case, the width peak B (in radians) was determined as full width at half-maximum (FWHM) by Gaussian fitting.

Particles morphology was investigated using Nova 200 NanoLab field emission scanning electron microscope (FE-SEM).

3. Neural networks modeling procedure

Artificial neural networks provide a mapping of inputs to outputs and consist of computer programs based on the structure of brain. As such, they can be trained to recognize patterns within data. In the human brain, a neuron is a nerve cell which processes incoming information and outputs a signal to the relevant part of the body accordingly. Some inputs are stronger than the others, i.e. they are 'weighted'. The total effect of the inputs is the sum of the weighted signals, and, if this exceeds the neuron threshold, a response is produced. By comparison, in an artificial neural network, a number of inputs are applied simultaneously, via weighted links, and the node calculates a combined total input. The relation between the input and output is specified by a transfer or activation function, which describes the threshold for deciding on the state of the output of that particular node. A number of nodes may be combined to form a layer, and layers may be interconnected to form a complete network. The procedure of designing the neural network architecture is described in detail as follows.

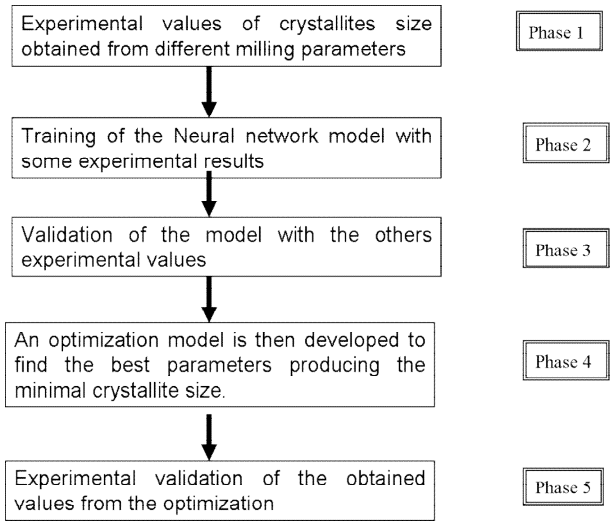


Fig. 1. Flowchart of the different steps.

4. Results and discussion

Fig. 1 summarized the different steps used in this study. A series of samples were prepared by using different milling parameters and the X-ray diffraction were used to determine the crystalline size. The process parameters, including milling times and ball-to-powder weight ratio are applied to the neural network inputs to provide information relating to particles size. The network is then trained to output the prediction on the powders particles size. An optimization model is then developed to find the best parameters producing the minimal average grain size and the last steps were the experimental validation.

Fig. 2 represents the evolution of XRD patterns of ZnO powder for the samples prepared under different milling condition. The pattern of un-milled ZnO powder shows a series of strong and narrow peaks characteristic for high quality ZnO crystals. With increasing the milling time, the diffraction peaks became broader and their relative intensity decreases. This effect is typical behavior of materials after milling and attributed usually to the presence of particles with small size and internal strain induced by mechanical deformation. In addition, all Bragg peaks of the XRD patterns showed only the ZnO reflections, indicating that there is no phase transformation during the milling have occurred. Table 1 summarized the particle size values calculated by Scherrer formula for all

Table 1. Experimental Particle size obtained by Scherer formula.

Balls to powder ratio: R	Times milling (h): T	Particles size (nm): PS
10	1	104
	3	98
	5	95
15	1	95
	3	69
	5	66
20	1	103
	3	93
	5	89

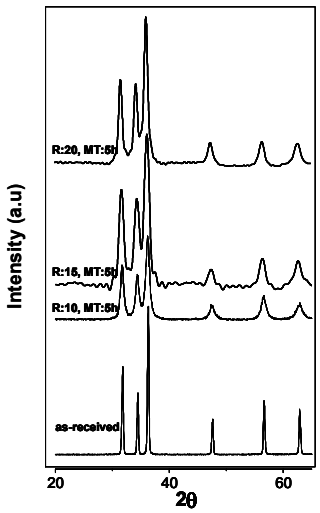


Fig. 2. X-ray diffraction patterns of ZnO prepared by milling under different condition.

samples.

Fig. 3 shows the evolution of crystalline size with milling time and the mass ratio of ball to powder obtained by Scherrer formula. For increasing milling time and for different mass ratio of ball to powder, the crystalline size decreases.

SEM micrographs of the samples before and after milling are shown in Fig. 4. It is clear that un-milled powder shows an inhomogeneities regarding particle size distribution (Fig. 4(a)). After one hour milling and for different mass ratio of balls to powder, a reduction of the particle size can be observed with relatively better homogeneity (Figs. 4(b) and 4(c)). It is clear that large particles are in fact agglomerates of much smaller particles.

An artificial neural network simulator has been developed to find out the relationship between the experience's inputs (the mass ratio of ball to powder and the milling time) and the experience's output (the average particles size). A multi-layer perceptron with backpropagation training has been implemented on MATLAB. The network is composed of 3 layers. There are 6 nodes in the input layer corresponding respectively to Ratio (R), Time (T), R*R, T*T, R*T, and the constant coefficient, which is set equals to one. There is only one node in the output layer corresponding to the particles size (S). There is one hidden layer composed of 6 nodes. The tanh is used as activation function for the hidden layer. Fig. 5 illustrate this network's architecture.

This artificial neural network was trained using the retro-propagation algorithm that minimizes the mean squared error

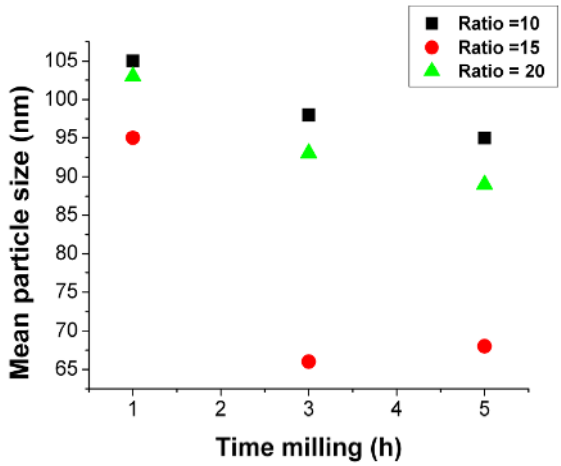


Fig. 3. Particles size as function milling time and mass ratio of ball to powder.

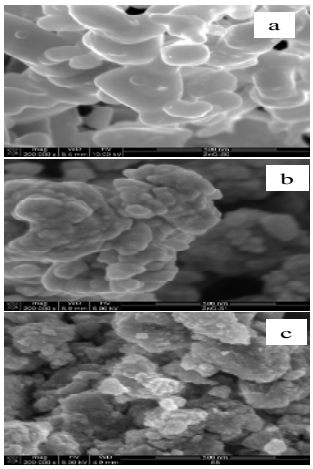


Fig. 4. SEM micrographs of ZnO powder: (a) as -received; (b) one hour milling and ratio = 10, (c) one hour milling and ratio = 15.

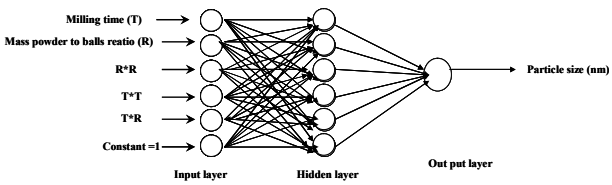


Fig. 5. The artificial neural network.

Table 2. Comparison between neural network and linear regression.

Experimental particles size (nm)	Network estimations	Regression estimation
104	103.9996094	110.161
98	98.00000517	97.409
95	95.00006589	95.321
95	95.00006589	91.536
69	68.99986047	77.534
66	65.99964682	74.196
103	102.9999497	114.561
93	93.00006064	99.309
89	88.99999527	94.721

over a training set of 8 experiences as given in Table 2.

This training algorithm was efficient. In few seconds, the Mean squared error has reached almost zero. The steady state is given by two matrixes. The first 2-dimension matrix, representing the input weights that connect the input layer to the hidden layer. The second 1-dimension matrix, representing the hidden weights that connect the hidden layer to the output node.

The particle size is then given by the following formula:

$$PS_{net} = \sum_{j=1}^{6}\left[wh_j \tanh\left(\sum_{i=1}^{6} x_i wi_{ij.} \right)\right]$$

where w_i and w_h are respectively the input and hidden wheights. The inputs xi are R, T, R*R, T*T, R*T, and 1. The obtained value is the multiplied by the standard deviation and added to the mean, as input data were normalized in order to speed up the training algorithm.

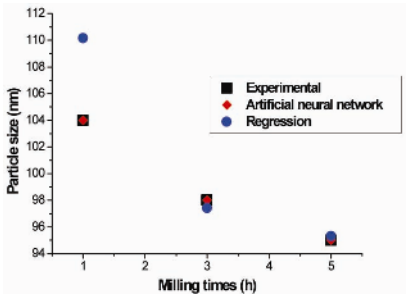
In order to assess the validity of the networks and their accuracy, it is often useful to perform regression analysis between the network response and the corresponding target.

In fact, using MINITAB software, we find the following multiple regression coefficients:

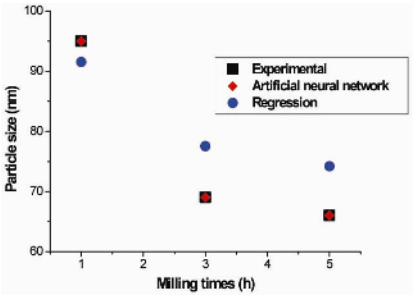
$$\mathrm{PS_{reg}} = 281.5 - 24.4R - 10.5T + .8R^2 + 1.3T^2 - 0.1RT$$

The mean absolute deviation and the mean absolute percent error obtained with this regression are respectively 5.65 and 6.64%. Our artificial neural network was very effective providing a perfect link between the inputs and the outputs, with an error by far better than multiple linear regressions.

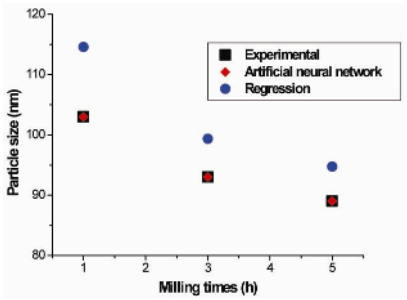
Table 2 provides a comparison between the performances of Regression and the multi-layer network described above. Fig. 6 shows that there is a consistence agreement between the outcomes of ANN modeling and experimental results as well as the current knowledge of mechanical milling process exists.



(a)



(b)



(c)

Fig. 6. Comparison between the performances of linear regression and artificial neural network: (a) balls to powder ratio: R = 10; (b) balls to powder ratio: R = 15; (c) balls to powder ratio: R = 20.

This agreement between the outcomes of ANN and experiments was obtained by Dashtbayazi et al. [18] and J. Maa et al. [19] but in our model the relative errors was better. The difference in the errors between ours models can be explained by the numbers of output parameters. In our model we were only interested by the crystallite size parameters. For their models the output was strain and crystallite size.

After constructing the ANN model and evaluating its accuracy and modeling error by regression analysis and total error estimation of the ANN network, one can use this network for prediction and optimization of the planetary mechanical milling process for synthesizing of ZnO nanopowders. A LINGO model is then developed to find the optimal ratio and milling time. The global optimal solution is found after a total of 2293 solver iterations. The minimal size is 61. The optimal values are 15.82832 for the ratio and 8.5 for the milling time. Further milling experiment is implemented using the above analysis results.

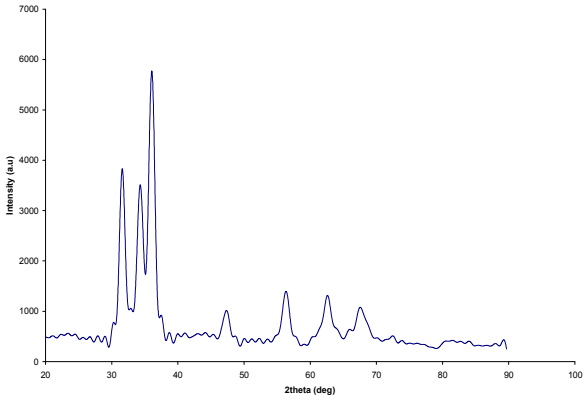


Fig. 7. X-ray diffraction of the experimental validation of the optimization model.

5. Experimental validation

According to the optimization model a time milling of 8.5 hours and balls to powder ratio of 15.8 are the best milling parameters that can produce a minimum crystalline size. A new milling experiment is implanted using these milling parameters. X-ray diffraction pattern is shown in Fig. 7. By using Scherrer formula, the crystalline size is about 63 nm. This value is close to that predicted by the optimization mode (61 nm).

6. Conclusions

An artificial-neural-network was developed to estimate the average crystalline size as a function on the milling parameters. The input parameters of the neural network are milling time and ball-to-powder weight ratio. It was found that the results from the neural network prediction perform a good coherence with the experimental data. The viability of the model is confirmed by the network prediction errors analysis. An optimization model and this experimental validation of the ball milling process for producing the nanopowder ZnO are carried out.

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Nomenclature

XRD : X-ray diffraction
SEM : Scanning electron microscopy
ANN : Artificial neural networks
PS : Particles size

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O. M. Lemine is is currently an Associate Professor at the physics department, College of Science, Al-imam University (Riaydh-Saudi Arabia). He received MSc (1995) and PhD (1999) in Materials Science from Henri Poicarre University (Nancy, France). His researches interests are in nd their applications including nanopowders and thin film. Before joining Al imam University, Dr Lemine was occupying several positions in Nancy University (France), Picardie University (France) and King Khaled University (Saudi Arabia).

M. A. Louly is currently a Professor at the Industrial Engineering Department, College of Engineering, King Saud University. He holds a Ph.D. in Operations Research and Management Science (UTT, Troyes, France), a Master in Computer Science (UHP, Nancy, France), and a Master in Mathematical Engineering (UHP, Nancy, France). His research focuses on Operations Research, Industrial Engineering, Production Planning, Transportations Problems, and Supply Chain Management. His main results are based on the exact mathematical programming methods and their intelligent coupling with heuristics algorithms. Before joining KSU, Prof Louly was occupying an associate professor position in the Industrial Engineering Department of University of technology of Troyes (France).