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Improving TiO_2 Nano Powder Synthesis Using High-energy Milling and Neural Network Optimization

Sumanth Ratna Kandavalli[1], Chandravathi Chandrasekaran[2], Amuthakkannan Rajakannu[3], Vimalanathan Palanikumar[4], Srinivasan Raju[5*], Venkatesan Srinivasan[6] and Mayakannan Selvaraju[7*]

[1]*Department of Mechanical Engineering, Tandon School of Engineering, New York University, Brooklyn, New York, USA*

[2]*Department of Information Technology, Saveetha School of Engineering, Saveetha Institute of Medical and Technical Sciences (SIMATS), Tiruvallur, Chennai, Tamil Nadu, India*

3Department of Mechanical and Industrial Engineering , National University of Science and Technology, Muscat, Sultanate of Oman, Oman

[4]*Department of Robotics and Automation, Rajalakshmi Engineering College, Thandalam, Chennai, India*

[5]*Department of Mechanical Engineering, Sri Krishna College of Technology, Kovaipudur, Coimbatore, India*

[6]*Department of Mathematics, K.S.R. College of Engineering, Tiruchengode, India*

[7]*Department of Mechanical Engineering, Vidyaa Vikas College of Engineering and Technology, Tiruchengode, Namakkal, Tamil Nadu, India*

***Correspondence to:**
Srinivasan Raju
Department of Mechanical Engineering,
Sri Krishna College of Technology,
Kovaipudur, Coimbatore, India.
E-mail: srinivasan310186@gmail.com

Mayakannan Selvaraju
Department of Mechanical Engineering,
Vidyaa Vikas College of Engineering and Technology,
Tiruchengode, Namakkal, Tamil Nadu, India.
E-mail: kannanarchieves@gmail.com

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Abstract

Using an artificial neural network (ANN) model trained on milling factors like milling duration (T) and balls-to-powder ratio (R), the crystalline size of TiO_2 (PS) nano powder was predicted. High-energy mechanical milling was used to produce this nano powder, and the results of the experiments served as the essential training data. Characterization of the produced TiO_2 nanoparticles using X-ray diffraction (XRD). ANN were shown to be superior to multiple linear regressions, with ANN modelling findings agreeing with experimental results with no significant error. Ball milling is used to make TiO_2 nano powder, and this experiment verifies the model used to optimize the process.

Keywords

Optimization, Artificial neural network, Titanium, Powder synthesis, Milling

Introduction

Semiconductors with a crystal size of 100 nm or less are a relatively new kind of material. The optical and electrical properties differ greatly from bulk semiconductors, while sharing comparable short range structures [1]. Applications of nano effects and quantum like effects in photo catalysis, nonlinear optical characteristics, and photoconductivity have been investigated [2]. The usage of TiO_2 nanoparticles has been shown to be beneficial in a variety of contexts including solar energy conversion, photocatalysis, light-emitting materials, transparent UV protection coatings, chemical sensors, and more [3, 4]. There are a wide variety of chemical and physical processes for creating TiO_2 nanoparticles [5]. Methods such bubble electrospinning, direct chemical synthesis, ball milling, and hydrothermal are only a few examples [6, 7]. Mechanical milling is one of these synthetic techniques that has shown promise as an easy way to create nanocrystalline powders with the potential for producing huge amounts of materials.

Nano powder characteristics are sensitive to milling parameters such as time, ball-to-powder mass ratio, rotation speed, ball diameters, and so on [8]. Ball milling process modelling has attracted the attention of several researchers, who have used both mechanical and thermodynamic techniques to try to get a more holistic, atomic, and phenomenological level understanding of the phenomenon

[9]. Combining the statistical method with an ANN has become one of the most powerful modelling tools in recent years. It is applicable to simulations of correlations that defy physical model description [10]. Saving time and money on expensive experiments is a major benefit of ANN modelling. To foretell steel plate deformations, authors [11] employed an ANN. Similar methods were utilized by authors [12] to optimize forced draught cooling tower operating parameters and evaluate performance. Predicting the results of mechanical milling may be done with ANN. However, research into neural networks' potential usefulness in mechanical milling is scant [13, 14]. The process of mechanical alloying (MA) has been modelled using an ANN created [15]. The major goal of their research was to identify the properties of the as milled metallic particles that were impacted by the various ball milling settings. With the model's assistance, predictions about the powders properties may be formed. To mimic the MA procedure, authors [16, 17] investigated the use of ANN. For WC-18% MgO nanocomposite powders, ANN were used to model the effect of milling time, milling speed, and ball-to-powder weight ratio on the powder's physical characteristics. By applying their model to WC-MgO composite powders, it may be possible to foretell how such powders would behave throughout different milling procedures [18].

The focus of this study is on improving the ball milling based manufacturing of nanocrystalline TiO_2 using a neural network model. Neural networks may be trained to predict particle size from limited amounts of process data, like milling duration and ball-to-powder weight ratios. After being fed data on powder particle sizes, the network is subsequently trained to make predictions. Next, an optimization model is constructed to determine which inputs result in the smallest mean grain size.

Experimentation

Preparation

The SPEX 8000 mixer mill was loaded with commercially available TiO_2 powders (99.9% purity) and stainless steel balls (diameters of 12 mm and 6 mm, respectively). Milling times ranged from 2 to 6 h, while ball-to-powder mass ratios went from 12 to 24.

Structural characterization

XRD studies were carried out utilizing a Shimadzu diffractometer (θ-2θ) equipped with Cu-K radiation (λ = 1.5426 Å). XRD line broadening is known to be affected by PS and interior stresses.

The PS was determined using the Schererrer formula [19, 20]

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

The entire width at half maximum (B in radians) was determined using a Gaussian fit to get the peak width B.

Modeling process of neural networks

The inputs and outputs of ANN are computed using computer program inspired by neural network. Inputs can be given different levels of importance, or "weights." The neuron will respond if the combined weighted signals from its inputs are greater than a predetermined threshold value [21]. However, in a neural network, several inputs are combined at once via weighted connections, and the sum of these inputs is computed at each node. A transfer or activation function defines the boundary conditions for a node's output state, characterizing the connection between input and output. Networks are built by connecting layers, each of which is composed of several nodes. Below is a full explanation of the method used to create neural network architecture.

Results and Discussions

Figure 1 provides a summary of the methodology employed in this investigation. XRD was used to measure the crystalline size after a series of specimen were produced using variant milling factors. Particle size data is generated by feeding process parameters into a neural network, such as T and R.

The network is then educated to make educated guesses about the powder's particle size. The following phase is the development of an optimization model to determine which parameters provide the smallest average grain size, followed by experimental validation.

Changes in XRD patterns of TiO_2 powder for samples milled at different conditions are shown in figure 2. Unprocessed TiO_2 powder has a structure characteristic of high quality TiO_2 crystals: a series of sharp, narrow peaks. Over longer milling periods, the diffraction peaks spread out and weaken. Milled materials are prone to this effect because mechanical deformation creates internal strain and the presence of small particles. There was no phase transition during milling since all Bragg peaks showed just TiO_2 reflections in the XRD patterns. The value for particle size determined using the Scherrer formula are summarized for all samples and shown in table 1.

Figure 3 displays the relationship between milling time and ball-to-powder mass ratio, as calculated by Scherrer

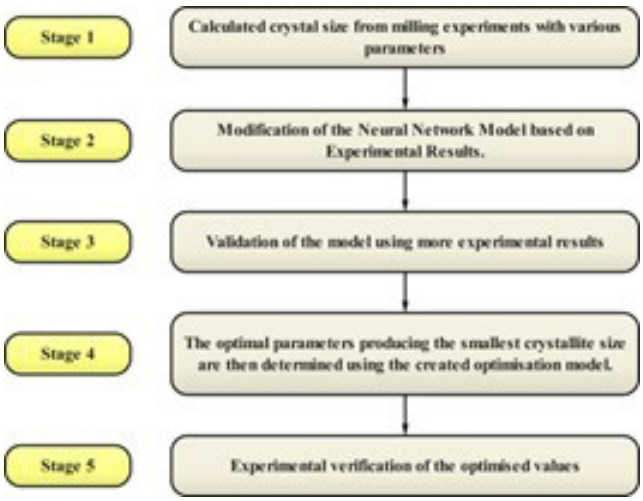

Figure 1: Diagram depicting the processes involved.

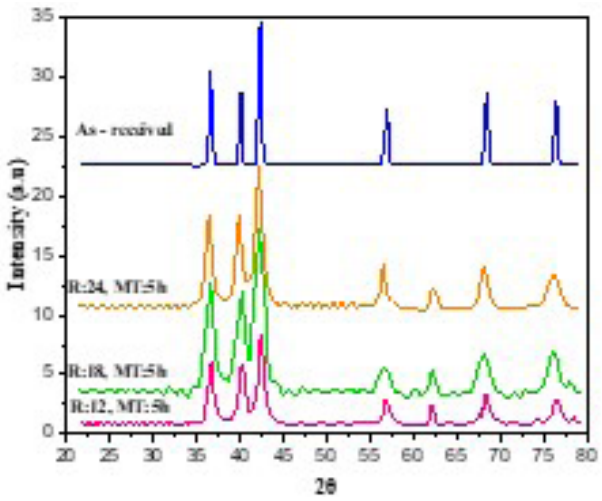

Figure 2: Milled TiO_2 with varying XRD patterns.

Table 1: Experimented size of particle calculated using the Scherrer formula.

Balls to powder ratio: R	Particles size (nm): PS	Times milling (h): T
12	113	2
	102	4
	99	6
18	99	2
	76	4
	73	6
24	112	2
	99	4
	92	6

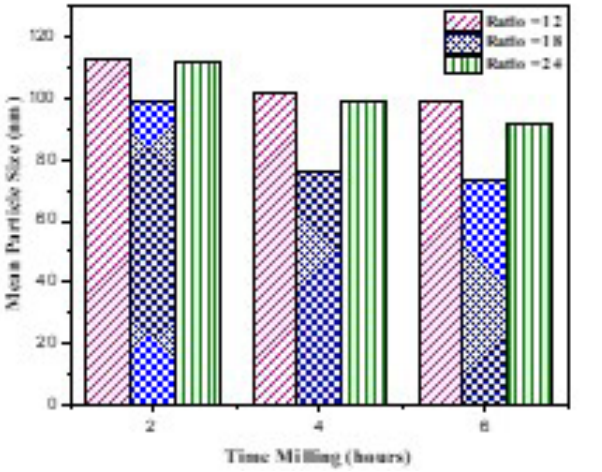

Figure 3: Milling time and ball-to-powder mass ratio as functions of particle size.

formula, and the subsequent growth in crystalline size. The size of the crystals is decreased when the time spent milling and the ball-to-powder mass ratio are adjusted.

The output of the experience may be predicted from the inputs using a neural network simulator. A back propagation-trained multi layer perceptron was built in MATLAB [22, 23]. The network consists of three distinct levels. The input layer consists of six nodes labelled "ratio," "time," "R*R," "T*T," "R*T," and "constant," which is always 1. In the final layer, the number of nodes represents the particle sizes (PS). One hidden layer with six vertices is present. The tanh is the secret layer's activation function. The layout of this network is seen in figure 4.

The mean squared error across the training set of eight encounters was minimized using the retro propagation strategy, as shown in table 2.

This training algorithm performed well. Mean squared error has nearly approached zero in a few seconds. Two matrices define the steady state condition. The input weightage from the input to the hidden layer, represented as a two-dimensional matrix initially. The second one-dimensional matrix represents the weights of the hidden connection between the hidden and the output layer.

The following formula yields the particle size [24, 25]: t

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

Where wi and wh are the input and hidden weightage, respectively. Since the training procedure was speed up by standardizing the input data, the final value is calculated by multiplying the standard deviation by the mean.

Regression analysis between a network's response and its associated target is a common method for gauging a network's validity and accuracy.

The multiple regression coefficients:

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

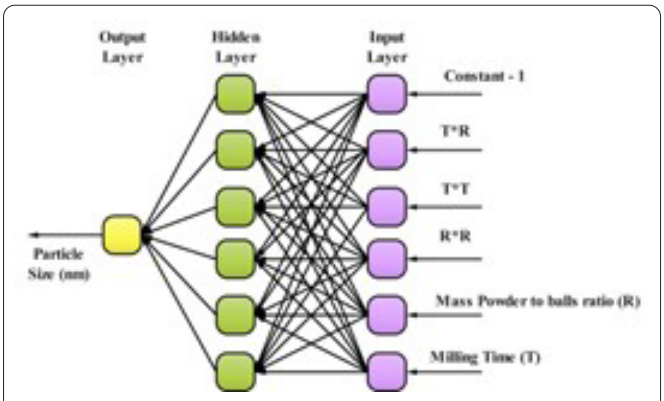

Figure 4: An ANN framework.

Table 2: Evaluating the neural network vs the linear regression.

PS (nm)	Network estimates	Regression estimates
113	113	119
102	102	101
99	99	99
99	99	96
76	76	85
73	73	81
112	112	124
99	99	105
92	92	98

The average percentage error and standard deviation from this regression are 5.65 and 6.64, respectively. When compared to several linear regressions, our ANN error rate was much lower since it established a perfect relationship between inputs and outputs.

The effectiveness of regression and the multi-layer network is compared in table 2. As can be shown in figure 5, there is agreement between the findings of ANN modelling and those of experiments, as well as with what is already known about the mechanical milling process. Authors [26] also found this agreement between the results of ANN and experiments; however, the relative errors in our model were less. The amount of output parameters explains the variation in error rates across our models. In our model, only the crystallite size characteristics were of importance. The result of their models was strain and crystallite size.

The planetary mechanical milling process for synthesizing TiO_2 nano powders may be predicted and optimized using the ANN network after its accuracy and modelling error have been tested by regression analysis and total error estimations. Next, the ideal milling ratio and duration are calculated using a LINGO model [27, 28]. After a total of 2293 iterations, the ideal global solution is discovered. The minimum quantity is 61. The ideal values for the ratio and milling duration are 15.82832 and 8.5, respectively. Using the aforesaid analytical results, more milling experiments are conducted.

Experimented Validation

The best milling factors for producing a minimal crystalline size, as determined by the optimization model, are 8.5 h of T and R of 15.8. A fresh milling study will be conducted with these values in mind. Figure 6 shows an example of an XRD pattern. The crystal's size is estimated to be 63 nm using Scherrer's formula. The anticipated value from the optimization method is quite like this one (61 nm).

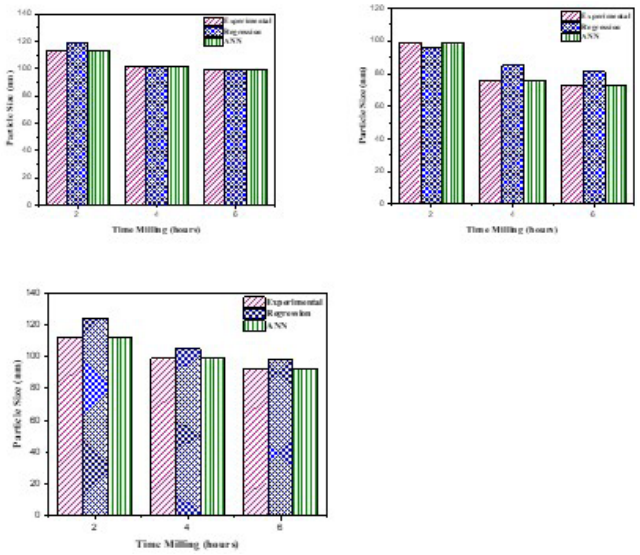

Figure 5: Evaluation the results of linear regression and ANN: **(a)** R = 12; **(b)** R = 18; and **(c)** R = 24.

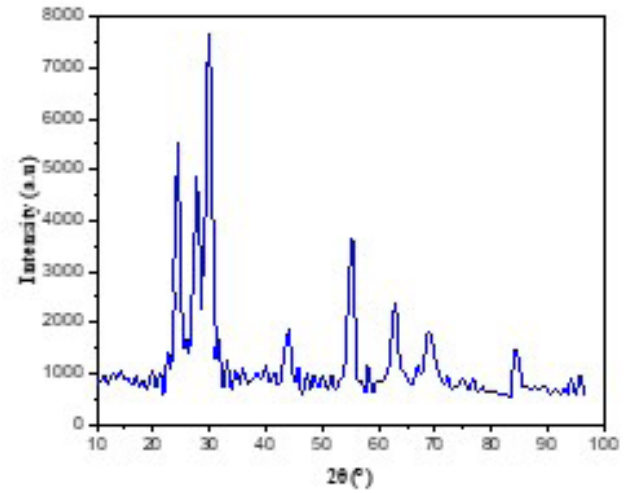

Figure 6: XRD of the experimented validation of the optimization model.

Conclusion

To determine how the milling conditions affect the average crystalline size, a neural network was developed. The neural network takes several factors into account, such as T and R. The results of the ANN predictions were found to agree with the data from the experiments. The model's viability is confirmed by an examination of the mistakes made by the network in its predictions. The ball milling method is experimentally verified and optimized to produce TiO_2 nano powder.

Acknowledgements

None.

Conflict of Interest

None.

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