Prediction of Silicon Direct Nitridation Kinetic By An Efficient and Simple Predictive Model Based on Group Method of Data Handling

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1. Introduction

Silicon nitride as one of the most practical engineering ceramics has been widely applied as a high temperature structural material. Several advantages such as high resistance against thermal shock and corrosion, high strength, lower density in comparison with metallic component, and biocompatibility make it popular as a structural material in cutting tools, gas turbine, diesel engine, precision bearings, and biomaterials. RBSN production method is one of the most economical methods to produce the silicon nitride for near net shape formation in comparison with the other producing methods. Although the RBSN method is cost-effective, it takes time for complete nitriding. Several parameters like physical properties of silicon pellet (e.g., size, density, pore size, and pore type) and gas properties (composition, flow rate, and pressure) can be effective on reactant diffusivity. A flexible model that can effectively consider all these parameters is one of the most important issues in simulation and prediction of kinetic behavior of silicon nitridation. Several efforts have been achieved to model the kinetic of reaction. Most of these models have been developed to illustrate gas-solid reactions (without presence of catalyst). Models are classified in three main groups: pore model, particle-pellet model and volume reaction model. The sharp interface model (SIM) has been introduced as a specific condition for gas–solid reactions for the cases that the primary particles are solid and nonporous. The main idea behind this assumption is that the reaction occurs in the sharp interface of reacted and unreacted solids while the reaction takes place in the whole pellet in case of the porous particles. SIM conditions could be applied to both particle-pellet and pore models.

In general, there are different mechanisms that control the kinetic of silicon nitridation during the reaction. Mechanisms involved at initial stage can be also completely different from the final stage. Furthermore, in special duration of reaction, several mechanisms may work in parallel. In this regard, the mentioned analytical models cannot consider all complex mechanisms involved during the reaction. As a result, there are always remarkable scatters between experimental results and the predictions of mentioned models during the whole reaction or a special period of reaction. On the other hand, several influencing parameters affect the reaction kinetic that must be considered in the mentioned models.

Recently, machine learning approaches such as Artificial Neural Networks (ANNs) and ANFIS as the most common soft computing methods have been employed to overcome these limitations in material science. Results confirmed the superior performance of data mining-based approaches. However, the ANN and ANFIS models do not give enough insight into the generated models and are not as easy to use as the empirical formulas. Among the soft computing methods, the GMDH network is known as a self-organized method to model and discover the behaviors of unknown or complicated systems based on given input–output data points. The main objective of this study is to investigate the efficiency of the GMDH network for predicting the kinetic of silicon nitridation. The main advantage of GMDH method in comparison with methods like ANN is that the dependencies between input parameters and output parameter are represented in parametric form as an equation while these dependencies are hidden within neural network structures in ANN method. To develop a simple and efficient predictive model based on GMDH, a comprehensive database from literature containing 2186 experimental results is applied. The developed GMDH model related the conversion percentage of silicon to the time, temperature, nitrogen percentage, pellet size, and silicon particle size. The developed GMDH results are also compared with two most common existing models through statistical error indicators. The relative importance of significant parameters dealing with conversion percentage is also investigated through sensitivity analysis. The robustness of proposed GMDH model is also verified through a parametric analysis.

2. Result and Discussions

2.1. Performance Analysis. The results of developed GMDH model and the actual experimental observations for training and testing datasets are depicted in Fig.1, respectively. Furthermore, the errors and the best fitted normal distribution on these errors are shown in these figures. As shown, there are good agreements between the predicted conversion percentage by GMDH model and the observed ones for both training and testing datasets.

To quantitatively evaluate the performance of developed model, the statistical error parameters are presented in Table 1 for both training and testing datasets. The comparisons between the results of Li et al. (1997) and Chang et al. (2000) models and the developed GMDH model are depicted in Fig. (2). According to this figure, in general, the developed GMDH model showed a remarkable agreement with experimental results for the whole duration of reaction and outperformed the other models.

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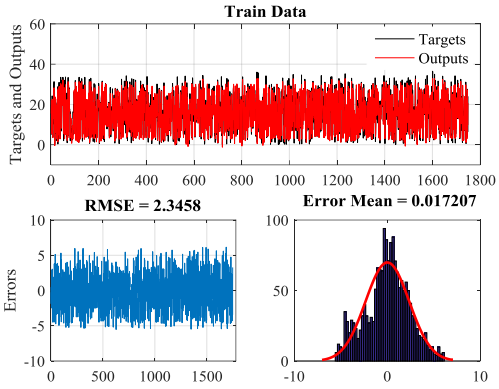

Fig. 1. Results of predicted model and experimental observations

Table 1Validation of the proposed model by Statistical error measures

Item	Formula	Condition	Training data set	Testing data set
BIAS	$Bias = \frac{\sum_{i=1}^{N}(P_i - O_i)}{N}$	$\lvert BIAS \rvert \to 0$	-0.0172	-0.1161
RMSE	$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)^2}$	$RMSE_{train}=RMSE_{test}$	2.3458	2.3827
R	$R = \frac{\sum_{i=1}^{N}(P_i - P_m)(O_i - O_m)}{\sqrt{(P_i - P_m)^2}\sqrt{(O_i - O_m)^2}}$	$R > 0.8$	0.9660	0.9654
R^2	$R^2 = 1 - \frac{\sum_{i=1}^{N}(O_i - P_i)^2}{\sum_{i=1}^{N}(O_i - O_m)^2}$	$R^2 \to 1$	0.9331	0.9317

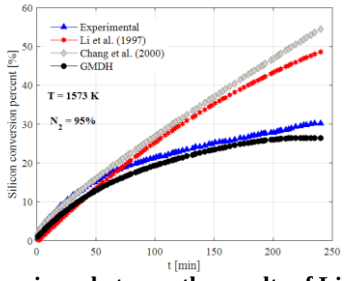

Fig. 2. comparisons between the results of Li et al. (1997) and Chang et al. (2000) and the developed GMDH models

2.2. Sensitivity Analysis

To determine the most important parameters in estimation of conversion percentage, a sensitivity analysis (ST) is done. To more illustrate the results of GT analysis, the values of V_{ratio} for different scenarios are shown in Fig. 3.

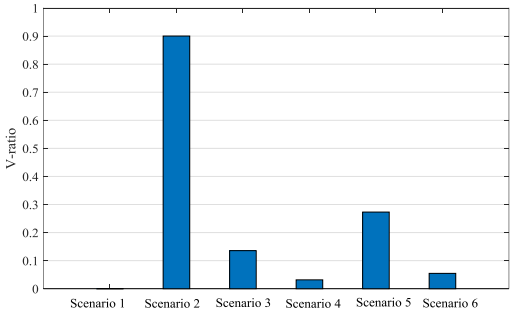

Fig 3. Results of Gamma Test (the values of Vratio for different scenarios)

2.3. Parametric Analysis

To more validate the robustness of developed model, a parametric analysis is performed to ensure that the results of proposed GMDH model are in line with physical concepts of the reaction kinetic. The results of parametric analysis are presented in Fig.4

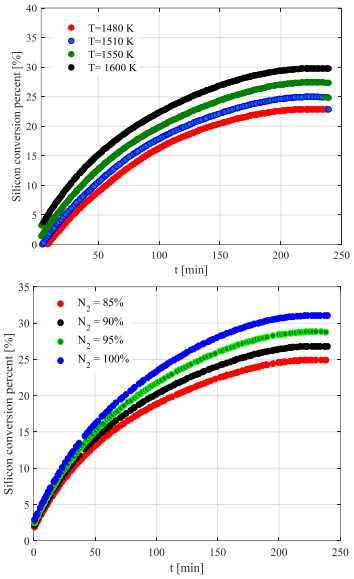

Fig. 4. The results of parametric analysis for a) different temperatures b) nitrogen percentage

3. Conclusions

In the present study, an effective predictive model based on group method of data handling (GMDH) is developed to estimate the kinetic of silicon nitride formations. To achieve this, a comprehensive database including 2186 data vectors is obtained from experimental results in literature. Several influencing parameters such as time, temperature, nitrogen percentage, pellet size, and silicon particle size are considered as input variables to model the conversion percentage. The accuracy of developed GMDH model is evaluated through the most common statistical error parameters. The results of performance analysis indicated that the developed model with R=0.966 and R^2=0.93 has a remarkable accuracy in estimation of conversion percentage. To further confirm the accuracy of developed model, its performance is also compared with two most common previous models of Chan et al. (2000) and Li et al. (1997). The results showed that the previous models perform well in earlier times of reaction while their predictions are remarkably overestimate after time of 75 min. However, the developed GMDH model showed a remarkable agreement with experimental results for the whole duration of reaction.

To determine the most effective parameters in prediction of conversion percentage, the Gamma Test (GT) is applied. Results of sensitivity analysis indicated that the time and nitrogen percentage are the most important parameters. The temperature, silicon particle size, and pellet size were the other important parameters, respectively. To ensure that the results of developed model are in line with physical concepts latent in the problem, the parametric analysis is also implemented. The results confirmed the robustness of GMDH model in capturing physical patterns in the used database and also previous studies.