

May A. Muslim

Department of Chemical
Engineering, University of
Technology, Baghdad, Iraq
mayrashid1973@gmail.com

Zainab Yousif

Department of Chemical
Engineering, University of
Technology, Baghdad, Iraq

Mohamed A. Abdel**Ghany**

Department of Chemical
Engineering, University of
Technology, Baghdad, Iraq

Bamidele V. Ayodele

Faculty of Chemical and
Natural Resources Engineering,
University Malaysia Pahang,
Lebuhraya Tun Razak, 26300
Gambang-Kuantan Malaysia

Received on: 30/09/2018

Accepted on: 10/01/2019

Published online: 25/04/2019

Predictive Modeling of Multilayer Graphene Growth by Chemical Vapour Deposition on Co-Ni/Al₂O₃ Substrate using Artificial Neural Network

Abstract- The uniqueness of multilayer graphene as extremely high carrier mobility, tune-able band gap and high elasticity has made it be considered as a high prospect engineering material that can be employed for several applications such as solar cells, field effect transistors, super-capacitors, batteries and sensors. In this study, the application of Artificial Neural Networks (ANN) for the predictive modeling of multilayer graphene (MLG) growth by chemical vapor deposition (CVD) on Co-Ni/Al₂O₃ substrate was investigated. Data comprises temperature, catalyst compositions, ethanol flowrates were generated using central composite experimental design and employed to obtain the MLG yield as the response. The data were subsequently used for predictive modeling using ANN. The findings show that the predictive values of the MLG yields were in good agreement with those obtained from the experimental runs having a coefficient of determination (R^2) of 0.988.

Keywords- Artificial Neural Network, Chemical Vapour Deposition, Multilayer Graphene, Predictive modeling, Cobalt-Nickel substrate.

How to cite this article: M.A. Muslim, Z. Yousif, M.A. Abdel Ghany and B.V. Ayodele "Predictive Modeling of Multilayer Graphene Growth by Chemical Vapour Deposition on Co-Ni/Al₂O₃ Substrate using Artificial Neural Network," *Engineering and Technology Journal*, Vol. 37, Part C, No. 1, pp. 113-119, 2019.

1. Introduction

The advent of nanotechnology and quest for miniaturization of electronic components in scientific equipment has aroused research interest in graphene and multilayer graphene [1]. The research interest in graphene and multilayer graphene as important engineering materials can be attributed to their excellent electrical, thermal, mechanical properties [1–3]. Also, graphene and multilayer graphene displayed specific interesting properties in transport phenomena, high Young's modulus, fracture strength, thermal conductivity, specific surface area and chemical stability [4,5]. Generally, graphene possesses a two-dimensional structure with a sp²-hybridized carbon atoms between one and ten layers arranged in a six-membered ring [6]. The physicochemical properties of graphene and multilayer graphene are often influenced by the preparation method [7]. Up to date, several techniques such as chemical vapour deposition, epitaxial growth, arc discharge, unzipping of carbon nanotube,

electrochemical synthesis, total organic synthesis, chemical reduction of graphene oxide, and plasma discharge etching of graphite have been employed for the synthesis of graphene and multilayer graphene [4,6,8,9]. Amongst these techniques, chemical vapour deposition has been reported has suitable techniques to synthesized graphene and multilayer graphene in a commercial quantity [10,11]. The chemical vapour deposition method has been employed to produce graphene and multilayer graphene with excellent electrical and optical properties with large specific surface area [12]. Ali et al. [13] reported the synthesis of multilayer graphene by chemical vapour deposition on bimetallic Co-Ni/Al₂O₃ catalyst. The synthesized multilayer graphene was characterized by Scanning Electron Microscopy (SEM), Transmission Electron Microscopy, Energy Dispersive X-ray (EDX), Raman Spectroscopy, and X-Ray Diffraction analysis. The characterization analysis showed that the multilayer graphene possesses excellent physicochemical properties for gas sensing

<https://doi.org/10.30684/eti.37.1C.18>

2412-0758/University of Technology-Iraq, Baghdad, Iraq

applications. In a follow-up study, the multilayer graphene was employed as the material for the ammonia sensor [14]. The study revealed that the multilayer graphene has a high sensitivity to ammonia sensing. In the present study, the predictive modeling of multilayer graphene growth by chemical vapour deposition over Co-Ni/Al₂O₃ using the artificial neural network is being investigated. The use of the artificial neural network in the engineering materials manufacturing industries is receiving growing attention due to its ability to predict process with complex non-linear relationship [15]. Due to its robust performance, ANN approach has been employed to predict the mechanical characterization of hydrogen functionalized graphene sheets [16]. The study shows that ANN excellently predicted the tensile strength of the hydrogen functionalized graphene sheet. The predicted property was consistent with that obtained from the experimental run. In the present study, the ANN techniques have been applied for the prediction of multilayer graphene yield synthesized using chemical vapour deposition method considering input parameters such as CVD temperature, Co/Ni ratio, and ethanol flowrate.

2. Materials and Methods

I. Synthesis and Characterization of the multilayer graphene

The detail of the synthesis and characterization of the multilayer graphene has been reported in Ali et al. [13,17]. Briefly, Al₂O₃ weighing 0.61-0.67 was functionalized via immersion in a mixture consisting of 50 ml of HNO₃ solution, 5 ml H₂SO₄, and 500 ml of de-ionize water. The mixture was agitated for 30 min in a mechanical shaker. After that, the Al₂O₃ was left in the mixture for one week to allow surface etching for the deposition of Co-Ni catalyst. The Co and Ni precursors Ni (NO₃)₂.6H₂O and Co (NO₃)₂.6 H₂O were mixed in different proportions in 50 ml of ethanol and left for 24 h. The catalyst mixture was sonicated for enhancing dispersion on the Al₂O₃. The Co-Ni/Al₂O₃ was prepared by dropping 6 ml of the Co-Ni substrate mixture on the Al₂O₃ which was placed in a heater.

The multilayer graphene was synthesized by initially weighing a stipulated amount of the as-prepared Co-Ni/Al₂O₃ and placed it in a CVD furnace reactor using ethanol as the carbon source. Before the start of the synthesis of the multilayer graphene, the CVD reactor was purged

for 30 min using argon gas. The ethanol was a heater and the vapour was allowed to flow through the CVD reactor which temperature was varied between 700-800 °C during which the multilayer graphene film was deposited on the Co-Ni/Al₂O₃. The Co-Ni/Al₂O₃ and the multilayer graphene were characterized for their physicochemical properties as reported by Ali et al. [13]. The results showed that the Co-Ni/Al₂O₃ and the multilayer graphene possessed appropriate physicochemical properties for catalytic processes and gas sensing applications, respectively.

II. Artificial Neural Network Modeling of the multilayer graphene

The steps involve in the ANN predictive modeling of multilayer graphene yield is depicted in Figure 2. The detail of the steps is as follow:

III. Data Collection using Design of Experiment (DoE)

The data employed for the ANN predictive modeling of the MLG yield was obtained from the Design of Experiment summarized in Table 1. The experiment to obtain the data was designed using a three-level, three-factorial central composite design (CCD).

IV. Determination of Input Data, Target and initialize the weight

The input parameters investigated include reaction temperature, catalyst composition, and ethanol flow rate while the output parameter was the multilayer graphene yield. The previous study has shown that reaction temperature, catalyst composition, and the ethanol flow rate has a significant influence on multilayer graphene yield (the target) produced by chemical vapour deposition [18].

V. Determination of ANN architecture

The ANN architecture employed in this study is shown in Figure 2. The architecture consists of a three layer-layer feed-forward structure with backpropagation algorithm [19]. The ANN architecture consists of an input, hidden and output layers. There are three-point vectors in the input layers which are based on the input parameters while the output layer is made of a point vector which is also based on the output parameter.

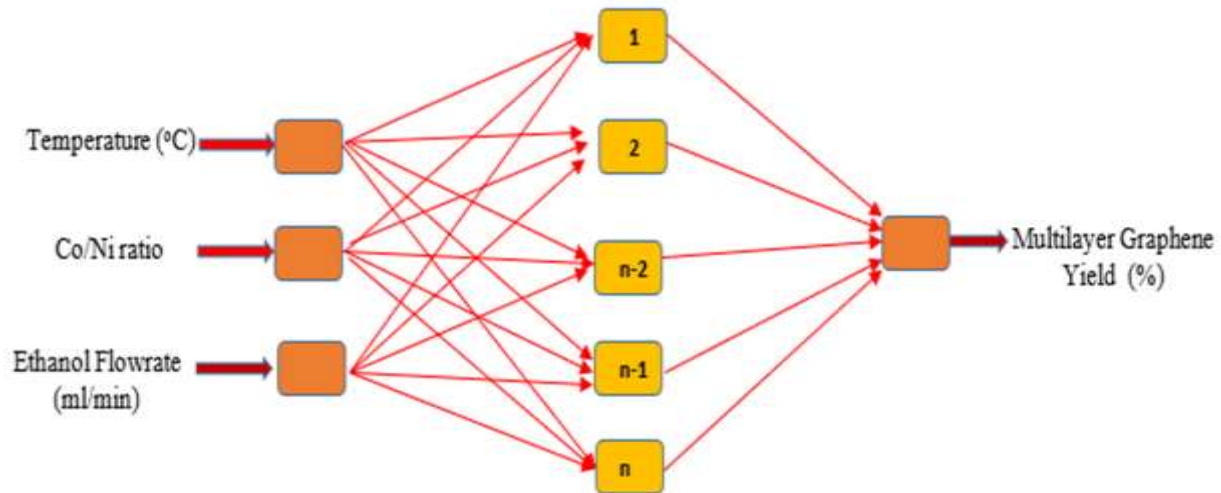


Figure 1: ANN architecture for the predictive modeling of the multilayer graphene.

VI. Training and Validation of the ANN architecture

The ANN architecture was trained to properly identified the linear and non-linear relationship between the input and output parameters. This is to enable the ANN to optimize the best weight and the input vectors suitable to give the best fit to the target vector. During the training, the input parameters were propagated through the network to determine a comparison between the error functions. The main objective of this procedure is to minimize the errors through the adjustment of the weight. The weight with the minimum error is employed to finally train and validate the ANN to obtain the best predictive output. The network was trained using the neural fitting tools in Matlab version 7.10 (2010a). A total of 20 elements (data set) was imputed into the neural network out which 70% was used for training, 15% for validation and 15% for testing of the neural network predictive model.

7. Results and Discussion

I. Optimization of the hidden neuron

The optimization results to minimize the network error to obtain the best-hidden neuron to train the network is shown in Table 1. To minimize the mean, standard error of the network, a series of hidden layers (at 2-interval) were employed to train the network [21]. The result of the optimization of the best-hidden neuron that minimizes the MSE is shown in Table 1. The analysis revealed that the network with 10 hidden neurons gives the least MSE of 1.27×10^{-13} with a perfect coefficient of determination (R^2). This

implies that the use of the 10 hidden neurons for subsequent training, validation and testing of the neural network will result in a minimum error with an accurate prediction of the multilayer graphene yield.

II. Comparison between the measured and the ANN predicted multilayer graphene yield

The parity plots showing the measured (experimental values) and the ANN predicted multilayer graphene yield obtained during the training, validation and the testing of the network are depicted in Figure 3 (a)-(c). To test the robustness of the ANN to predict the multilayer graphene yield, the fittings were subjected to statistical evaluation as shown in Table 2. Based on Figure 3, it can be seen that both the experimental values of the multilayer graphene and those of the ANN predicted values are close to the linear trendline for training, validation and the testing. This observation implies that both the experimental values of the multilayer graphene and the ANN predicted values are in close agreement. This is an attestation to the robustness and reliability of the ANN to predict multilayer graphene yield based on the linear and non-linear relationship between the input parameters and the target [20,22]. Further statistical analysis to evaluate the reliability of the ANN model was performed using the coefficient of correlation (R), coefficient of determination (R^2), standard error of estimate (SEE), and the p-value [23]. The R was employed to measure the degree of relationship between the input parameters and the target. The R values of 0.9846, 0.9946, and 0.9983 obtained for the network training,

validation and testing implies that there is a strong correlation between the input parameters (CVD temperature, Co/Ni ratio, ethanol flow rate) and the target (multilayer graphene yield). Also, the R^2 was employed to determine the percentage variation in the multilayer graphene yield which is explained by all the input parameters al together. The R^2 values of 0.9694, 0.9882, and 0.9966 obtained for the network training, validation and testing signifies that there was a little variation in the multilayer graphene

yield which is explained by all the input parameters. The standard error of estimate measures the accuracy of the ANN prediction of the multilayer graphene using the regression line. A slightly higher SEE of 1.556 was obtained during the network training. The SEE was gradually reduced to 0.999 and 0.522 during the network validation and testing which further strengthens the ability of ANN to accurately predict from a given dataset.

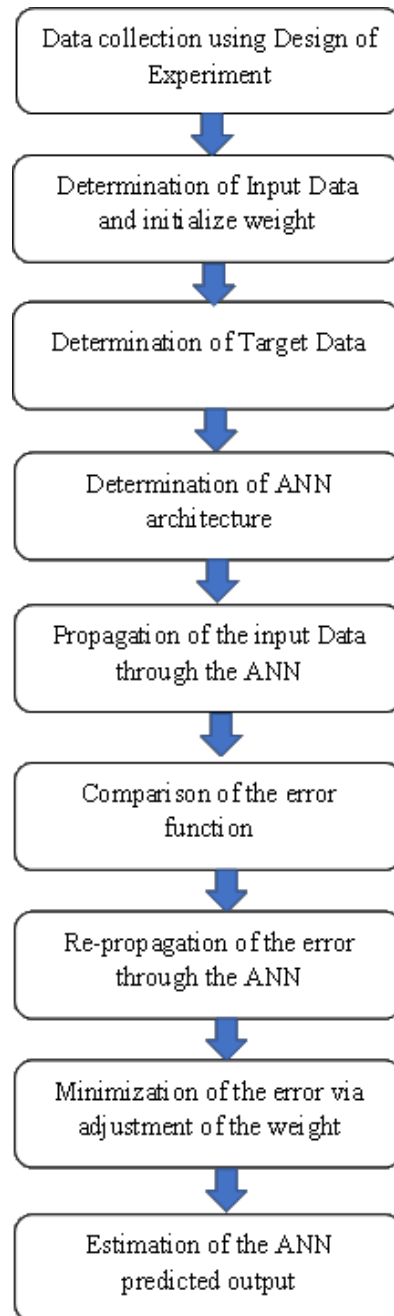


Figure 2: Steps involved in the ANN predictive modeling of MLG yield (Adapted from on all et al.

Table 1: Data employed for the ANN predictive modeling

Input parameters				Output parameters
Run	Reaction Temperature (°C)	Catalyst composition (%)	Ethanol flow rate (ml min ⁻¹)	MLG Yield (%)
1	700.00	0.30	9.00	71.00
2	800.00	0.30	9.00	70.70
3	700.00	0.70	9.00	54.00
4	800.00	0.70	9.00	62.50
5	700.00	0.30	11.00	75.00
6	800.00	0.30	11.00	75.10
7	700.00	0.70	11.00	60.20
8	800.00	0.70	11.00	63.20
9	665.91	0.50	10.00	59.10
10	834.09	0.50	10.00	65.20
11	750.00	0.16	10.00	70.20
12	750.00	0.84	10.00	45.30
13	750.00	0.50	8.32	70.90
14	750.00	0.50	11.00	75.50
15	750.00	0.50	10.00	75.71
16	750.00	0.50	10.00	77.00
17	750.00	0.50	10.00	76.00
18	750.00	0.50	10.00	74.00
19	750.00	0.50	10.00	75.00
20	750.00	0.50	10.00	73.00

Table 2: Optimization of the Hidden Neuron to obtain the least MSE

Number of Hidden Neuron	Mean Square Error (MSE)	Coefficient of Determination (R ²)
2	4.53x10 ⁻²	0.999
4	74.17	0.974
6	141.93	0.975
8	86.67	0.934
10	1.27x10 ⁻¹³	1.000
12	64.33	0.977
14	1.18x10 ⁻³	0.999
16	8.55x10 ⁻¹	0.994
18	3.88x10 ⁻⁵	0.999
20	189.56	0.877

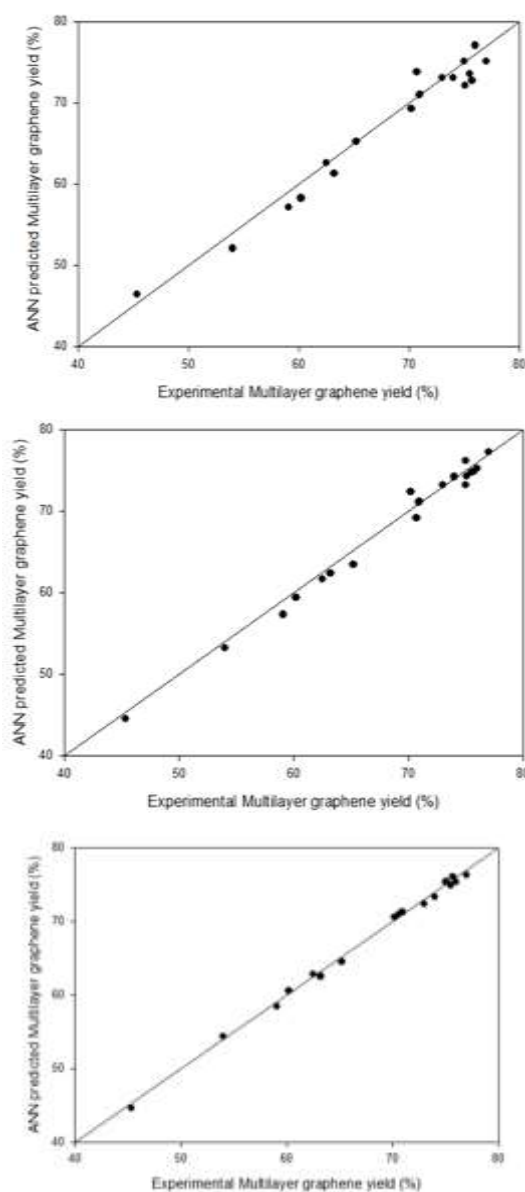


Figure 3: Parity plot showing the comparison between the experimental and ANN predicted multilayer graphene yield (a) Training (b) Validation (c) Testing

Table 3: Statistical analysis of the ANN predictive modeling

	R	R^2	Standard Error of Estimate	p-value
Training	0.9846	0.9694	1.5563	<0.0001
Validation	0.9941	0.9882	0.9990	<0.0001
Testing	0.9983	0.9966	0.522	<0.0001

4. Conclusion

A conclusion should point out the distinguished this study has demonstrated the use of ANN for the prediction of multilayer graphene yield. The ANN architecture which consists of three layer-layer feed-forward structure with back propagation algorithm revealed the accurate prediction of the multilayer graphene yield with high values of R (0.9941), R^2 (0.9882) and low value of the standard error of estimate (0.999). The comparison of the experimental values of the multilayer graphene yield and the ANN predicted values is consistent, an indication of the

reliability and the robustness of the ANN for the prediction of multilayer graphene yields. This study has demonstrated that ANN can be employed for the prediction of multilayer graphene yield in an industrial production scenario considering the relationship between the input parameters and the target.

5. Acknowledgment

The authors to appreciate the financial support of University of Technology, Baghdad, Iraq.

References

- [1] B. Tang, G. Hu, H. Gao, L. Hai, "Application of graphene as filler to improve thermal transport property of epoxy resin for thermal interface materials," *Int J Heat Mass Transf*, Vol. 85, pp. 420–429, 2015.
- [2] M.M. Sadeghi, M.T. Pettes, L. Shi, "Thermal transport in graphene," *Solid State Commun*, Vol. 152, pp. 1321–1330, 2012.
- [3] J.W. Suk, W.H. Lee, J. Lee, H. Chou, R.D. Piner, Y. Hao, D. Akinwande, R.S. Ruoff, "Enhancement of the electrical properties of graphene grown by chemical vapor deposition via controlling the effects of polymer residue," *Nano Lett*, Vol. 13, pp. 1462–1467, 2013.
- [4] E.-Y. Choi, T.H. Han, J. Hong, J.E. Kim, S.H. Lee, H.W. Kim, S.O. Kim, "Noncovalent functionalization of graphene with end-functional polymers," *J Mater Chem*, Vol. 20: 1907, 2010.
- [5] G. Jo, M. Choe, C.-Y. Cho, J.H. Kim, W. Park, S. Lee, W.K. Hong, T.W. Kim, S.J. Park, B.H. Hong, Y.H. Kahng, "Large-scale patterned multi-layer graphene films as transparent conducting electrodes for GaN light-emitting diodes," *Nanotechnology*, Vol. 21, pp. 17520, 2010.
- [6] K. Lü, G.X. Zhao, X.K. Wang, "A brief review of graphene-based material synthesis and its application in environmental pollution management," *Chinese Sci Bull*, Vol. 57, pp. 1223–1234, 2012.
- [7] S. Sheshmani, M.A. Fashapoyeh, "Suitable chemical methods for preparation of graphene oxide, graphene and surface functionalized graphene nanosheets," *Acta Chim Slov*, Vol. 60, pp. 813–825, 2013.
- [8] C.-M. Seah, S.-P. Chai, A.R. Mohamed, "Mechanisms of graphene growth by chemical vapour deposition on transition metals," *Carbon N Y*, Vol. 70, pp. 1–21, 2014.
- [9] B. Hu, H. Ago, Y. Ito, K. Kawahara, M. Tsuji, E. Magome, K. Sumitani, N. Mizuta, K.I. Ikeda, S. Mizuno, "Epitaxial growth of large-area single-layer graphene over Cu(1 1 1)/sapphire by atmospheric pressure CVD," *Carbon*, Vol. 50, pp. 57–65, 2012.
- [10] A.V. Tyurnina, H. Okuno, P. Pochet, J. Dijon, "CVD graphene recrystallization as a new route to tune graphene structure and properties," *Carbon*, Vol. 102, pp. 499–505, 2016.
- [11] P. Avouris, C. Dimitrakopoulos, "Graphene: synthesis and applications," *Mater Today*, Vol. 15, pp. 86–97, 2012.
- [12] C. Mattevi, H. Kim, M.A. Chhowalla, "A review of chemical vapour deposition of graphene on copper," *J Mater Chem*, 2011; Vol. 21, pp. 3324–3334, 2011.
- [13] M. Ali, S.A. Rashid, M.N. Hamidon, F. Md Yasin, "Facile Synthesis and Characterization of Multi-Layer Graphene Growth on Co-Ni Oxide/ Al_2O_3 Substrate Using Chemical Vapour Deposition," *Bull Chem React Eng Catal*, 2018; Vol. 13, pp. 341, 2018.
- [14] M. Ali, A.R. Rashi, M.N. Hamidos, M.F. Yasin, F.T. J. Al-Sudani, B.V. Ayodele, "Ammonia Sensing Using Multilayer Graphene Grown on Co-Ni/ Al_2O_3 Substrate," *I-SAMN* 2018, pp. 15–16, 2018.
- [15] P.G. Benardos, G.C. Vosniakos, "Optimizing feedforward artificial neural network architecture," *Eng Appl Artif Intell*, Vol. 20, pp. 365–382, 2007.
- [16] V. Vijayaraghavan, A. Garg, C.H. Wong, K. Tai, Y. Bhalerao, "Predicting the mechanical characteristics of hydrogen functionalized graphene sheets using artificial neural network approach," *J Nanostructure Chem*, Vol. 3, pp. 833–837, 2013.
- [17] M. Ali, S. Abdul-Rashid, M.N. Hamidon, F.M. Yasin, "The Growth Kinetics of Multilayer Graphene Grown on Al_2O_3 Substrate by Chemical Vapour Deposition," *Procedia Eng*, Vol. 148, pp. 1295–1302, 2016.
- [18] A. Hadi, J. Karimi-Sabet, S.M.A. Moosavian, S. Ghorbanian, "Optimization of graphene production by exfoliation of graphite in supercritical ethanol: A response surface methodology approach," *J Supercrit Fluids* 2016; Vol. 107, pp. 92–105, 2016.
- [19] P. Zahadat, J. Milewski, "Modeling electrical behavior of solid oxide electrolyzer cells by using artificial neural network," *Int J Hydrogen Energy*, Vol. 40, pp. 7246–7251, 2015.
- [20] D. Onalo, S. Adedigba, F. Khan, L.A. James, S. Butt, "Data driven model for sonic well log prediction," *J Pet Sci Eng* 2018; Vol. 170, pp. 1022–1037, 2018.
- [21] B.V. Ayodele, C.K. Cheng, "Modelling and optimization of syngas production from methane dry reforming over ceria-supported cobalt catalyst using artificial neural networks and Box-Behnken design," *J Ind Eng Chem* 2015; Vol. 32, pp. 246–258, 2016.
- [22] D.J. Scott, P.V. Coveney, J.A. Kilner, J.C.H. Rossiny, N.M.N. Alford, "Prediction of the functional properties of ceramic materials from composition using artificial neural networks," *J Eur Ceram Soc*, Vol. 27, pp. 4425–4435, 2007.
- [23] M.A. Hossain, B.V. Ayodele, C.K. Cheng, M.R. Khan, "Artificial neural network modeling of hydrogen-rich syngas production from methane dry reforming over novel Ni/ CaFe_2O_4 catalysts," *Int J Hydrogen Energy*, Vol. 41, No. 26, pp. 11119–11130, 2016.