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Modelling of Low-Temperature Sulphur Dioxide Removal Using Response Surface Methodology (RSM), Artificial Neural Network (ANN) and Adaptive Neuro-Fuzzy Inference System (ANFIS) †

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Abstract: Empirical and machine learning models are estimation tools relevant to obtaining scalable solutions to engineering problems. In this study, response surface methodology (RSM) was incorporated to correlate the experimental findings based on mathematical models. Artificial neural networks (ANN) and adaptive neuro-fuzzy inference systems (ANFIS) were the artificial intelligence tools used to create trainable algorithms. Feed data consolidated hydration temperature (50 to 90 °C), hydration time (3 to 7 h), sulphation temperature (120 to 160 °C), diatomite to hydrated lime ratio (0 to 1) and inlet gas concentration (500 to 2500 ppm) as the independent variables mapped against sulphur capture capacity (Y_1 —5 to 54%) and reagent utilisation (Y_2 —4 to 42%) as the dependent variables. Statistical error techniques such as root mean square (RMSE), mean square error (MSE), and the coefficient of determination (R^2) were used to quantify the model accuracy and cost analysis. The ANN models presented more acceptable and reliable predicted data, with R^2 values greater than 99% compared to the RSM and ANFIS models. The ANFIS models showed overfitting deficiencies that affected learning and training. These findings suggest that the ANN models are a more suitable option for accurate and dependable data estimation in similar engineering applications.

Keywords: desulphurisation; emission control; fuzzy inference systems; neural networks; numerical models

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1. Introduction

Coal is undoubtedly the primary raw material for electricity generation globally. South Africa is Africa's most developed economy and has high energy demands to sustain its vast industries and domestic energy appetite. A total of 13 state-owned Eskom power plants jointly generate 37,698 MW for consumption across South Africa [1]. This translates to Eskom being the leading sulphur dioxide (SO_2) emitter in Africa. SO_2 emissions have exacerbated human health through occurrences of respiratory or heart infections. Acid rain formation from the SO_2 in the atmosphere chemically combining with rainwater leads to the poisoning of aquatic ecosystems and damage to limestone, marble and cultural buildings [2,3]. Flue gas desulphurisation (FGD) is the standard approach relevant to mitigating SO_2 emissions. This end-of-pipe technique relies on an alkaline reagent that neutralises the acidic SO_2 compound in the flue gas stream, forming a sulphate (SO_4^{2-}) or sulphite (SO_3^{2-}) salt. Dry FGD is a system developed to counter the high start-up and maintenance costs associated with wet and semi-dry units. It is commercially and operationally limited by low SO_2 capture capacity and sorbent utilisation.

Enhancing the effectiveness and productivity of the dry FGD system involves the use of process simulation and optimisation techniques. Response surface methodology (RSM) is a quantitative method used to decipher the complex correlation between multiple independent parameters and dependent (response) variables. A set of mathematical equations based on different experimental architectures are generated to model this relationship. The experimental model can be anchored to either a Full Factorial Design (2-level or 3-level FFD), a Box Beknhen Design (BBD) or a Central Composite Design (CCD) [4]. An artificial neural network (ANN) is an artificial intelligence system used to map relationships in vast datasets through learning and extracting relevant features. This models the pathways in the human brain and how neuron signals travel through the brain cells to elicit a reaction. ANN uses sets of algorithms to create models that can be used to solve complex problems. It is made of three layers through which metadata traverses before an estimated output is produced [5]. The adaptive neuro-fuzzy inference system (ANFIS) is a hybrid soft computing technique that combines the learning ability of the ANN and fuzzy logic reasoning. ANFIS is sectioned into five layers, including the input layer, the membership function (MF) layer, the normalised layer, the fuzzy IF-THEN rule layer and the output layer. The MF permits tuning as a measure to improve computation accuracy [6].

In this study RSM, ANN and ANFIS models were integrated to dry FGD forecasting by statistically analysing the synthetic outputs generated and the error produced by each tool. We fully acknowledge that the experimental metadata used does not constitute a huge dataset. However, this is sufficient to create models that can be used as a blueprint in similar studies. The modelling results from this case study are relevant to establishing optimisation and simulation procedures applicable to dry FGD. This report attempts to fill in the gap left by the paucity of comparative literature on these three modelling techniques by offering useful details on the approaches that are most useful for predicting desulphurisation and sorbent conversion efficiency and how their performance compares against one another.

2. Materials and Methods

The sorbent synthesis procedure and the fixed bed SO_2 adsorption experiments occurred as documented in a previous study [7]. A unique sorbent was formulated using cellulose nanocrystals as the $Ca(OH)_2$ particle growth template, while diatomite was incorporated as the sorbent additive at different ratios. The polynomial model established in the RSM system was formulated using the Design Expert (v13.0.5.0) CCD, set at an alpha level of 2 ($\alpha = 2$). This generated 50 test runs from five inputs (hydration temperature—50 to 90 °C, hydration time—3 to 7 h, sulphation temperature—120 to 160 °C, diatomite to hydrated lime ratio—0 to 1) and two outputs (sulphur capture capacity (Y_1)—5 to 54% and reagent utilisation (Y_2)—4 to 42%). These metadata were used as feed information for the machine learning (ML) models (Table 1). The MATLAB R2015 v8.5.0.197613 language was used to initialise the ANN and ANFIS scripts. Prior to ML computation, metadata preprocessing was performed using the min-max normalisation procedure to minimise model inaccuracy that can arise due to differences in varable magnitude [8]. Several ANN models were trained using the Levenberg-Marquardt (LM), Bayesian Regularization (BR), and Scaled Conjugate Gradient (SCG) techniques, with hidden cell configurations ranging from 7 to 10. The linear *(purelin)* and nonlinear sigmoid *(logsig and tansig)* functions were investigated as trigger mechanisms for data transmission from the hidden layer to the output layer. The preset *dividerand* function randomly sectioned feed data into 70% training (30 sets), 15% testing (10 sets) and 15% (10 sets) validation. The ANFIS models involved the development of a grid partitioning (genfis1) or subtractive clustering (genfis2) Sugeno-type fuzzy inference system (FIS), where five inputs were classified with one output at a time. The *gbellmf*, *trapmf*, *trimf*, *gauss2mf*, *dsigmf*, *pimf*, and *gaussmf* membership functions were compared for the computation of the input data. 35 data sets were used to train the ANFIS models while 15 data sets were used in testing. The predicted values were analysed by the constant type MF after optimisation using either the hybrid or backpropagation algorithms. Model validation induced the application of accuracy evaluation using the R^2 method and the loss function analysis using RMSE and MSE

Table 1. Generic equations of the best RSM, ANN and ANFIS models.

Tool	Model or Algorithm	Equation
RSM	Quadratic model	$Y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{12} x_1 x_2 + \varepsilon$
ANN	BR algorithm	$Y = f(w^* x + b)$
ANFIS	Genfis1—BP algorithm	$Y = w_0 + w_1 x_1 + w_2 x_2 + \cdots + w_n x_n$

* β_0 is the intercept term, β_1 and β_2 are linear coefficient terms, β_{11} and β_{22} are the quadratic terms, x, x_1 and x_2 are the input terms, ε is the RSM error value, b is the ANN bias vector, w is the ANN synaptic weight, f is the node trigger function, $w_0, w_1 \cdots w_n$ are the ANFIS network weights and Y is the overall predicted data.

3. Results and Discussion

The models used in this study were assessed based on the forecasting accuracy of the sulphation (Y_1) and reagent utilisation (Y_2) responses. Conventionally, the superior model generates values in proximity to the actual values. RSM was investigated as a tool efficient in deciphering the relationship between intermingling multiple input parameters and expected targets. From the developed model, it is possible to identify the optimised input features that present a highly desired response. The RSM program developed a quadratic mathematical model with R^2 values of 0.9583 for Y_1 and 0.9614 for Y_2. The ANOVA data of both models indicated p-values of less than 0.0001 which validates their significance [9]. ANN utilised different model designs based on the number of hidden cells and the learning algorithm. The BR script combined with a 5-10-2 ANN design achieved an overall R^2 of 0.9973 with an MSE of 0.023. This results from enhanced learning as no validation steps are performed in the BR algorithm sectioning more data for network training. This algorithm can accommodate inputs from small data sets using a penalty term that minimises overfitting ensuing from poor data generalisation [10]. This feature renders this script relevant in mapping the feed data in our study. LM was investigated as the study constituted a regression problem. It also lacks computation complexity and is suitable when fast simulations are needed. SCG is also applicable to nonlinear and complex relationships which is present in the dataset used in this study. The SCG and LM were outperformed as they work effectively with large networks constituting vast metadata. 10 hidden cells had higher accuracy due to improved classification of unseen data, consequently, more patterns between the inputs and outputs could be represented by this architecture. The *logsig* activation function was adequate in adapting to occurrences of non-linearity in the feed data owing to its smooth S-shaped orientation. This permits an ANN system to comprehend more complex information within a data set thereby minimising overfitting [11]. All ANFIS models produced outcomes that performed poorly compared to the RSM and ANN models. The grid partitioning (genfis1) FIS attained the highest R^2 of 0.8988 when the *pi* MF was applied and optimised using the backpropagation algorithm. The genfis 1 FIS integrates a uniform grid to model the membership functions which improves the computation of unstructured data better than genfis 2. The *pimf* outperformed all other MF due to higher adherence to linear and non-linear variables. This is made possible by its bell-shaped model, which has a symmetric centre point that can map the correlation in a dataset with a complex variable design. The poor performance of the ANFIS models can be ascribed to overfitting. The performance of the RSM and ML models used to predict the Y_1 and Y_2 responses is graphically summarised in Figure 1.

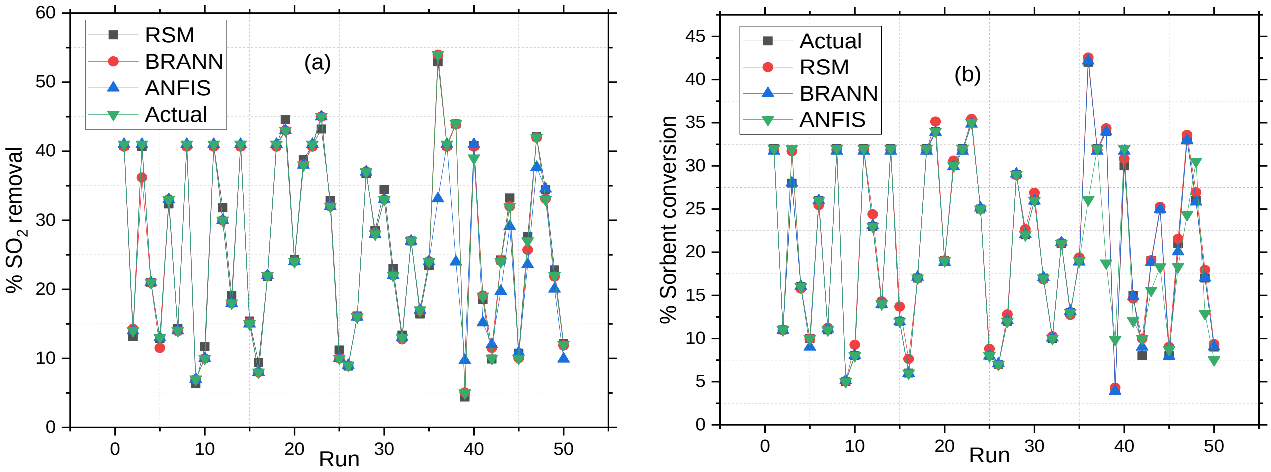

Figure 1. RSM, ANN and ANFIS synthetic data compared to the actual values of (**a**) SO_2 capture and (**b**) sorbent conversion.

The statistical data from the comparison of the forecasted and actual data is presented in Table 2. This data verifies the effectiveness of the ANN in creating models relevant to the dry FGD process.

Table 2. Error analysis of the RSM, ANN and ANFIS models.

Analysis	Sulphation Efficiency (%)			Sorbent Conversion (%)		
	RSM	BR	ANFIS	RSM	BR	ANFIS
RMSE	0.833334	0.4633	0.7056	0.68725	0.368223	0.5354
MSE	0.69446	0.2146	0.4979	0.47231	0.135588	0.2867
R^2	0.9753	0.9987	0.8988	0.9771	0.9986	0.8927

4. Conclusions

This work was intended to classify the forecasting performance of mathematical and ML models in low-temperature dry FGD. The results of this study can be applied in design and operation optimisation in the case of retrofit applications. This intends to significantly improve innovation and sustainability by ensuring reduced sorbent feed and high SO_2 removal. Data mapping involved using the RSM quadratic model and deep learning algorithms in the ANN and ANFIS tools. According to the statistical analysis, the 5-10-2 ANN layout surpassed the RSM and ANFIS models in computation accuracy with an R^2 of 0.9987 for Y_1 and 0.9986 for Y_2. This was achieved when simulations were done using the BR algorithm. The ANFIS script struggled with model overfitting even though it achieved high R^2 values (Y_1—0.8988, Y_2—0.8927), which may be related to a lack of training data needed to accurately train the network. This problem is being investigated in a future study by evaluating the use of cross-validation, early stopping technique and reducing the number of active neurons during training. It is crucial to remember that the findings of this research depend on the specific experimental setup and data collected. To verify the robustness and usability of the generated models, future work should seek to validate these findings using larger datasets and under other experimental situations.

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Abbreviations

ANOVA	Analysis of variance.
dsigmf	Difference between two sigmoidal membership functions
gbellmf	Generalised bell-shaped membership function.
gaussmf	Gaussian membership function.
logsig	Sigmoid activation function.
pimf	Pi-shaped membership function
purelin	Linear activation function
tansig	Hyperbolic tangent activation function.
trimf	Triangular membership function
trapmf	Trapezoidal membership function

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