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Prediction of gross calorific value of coal based on proximate analysis using multiple linear regression and artificial neural networks

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Abstract: Gross calorific value (GCV) of coal was predicted by using as-received basis proximate analysis data. Two main objectives of the study were to develop prediction models for GCV using proximate analysis variables and to reveal the distinct predictors of GCV. Multiple linear regression (MLR) and artifcial neural network (ANN) (multilayer perceptron MLP, general regression neural network GRNN, and radial basis function neural network RBFNN) methods were applied to the developed 11 models created by different combinations of the predictor variables. By conducting 10-fold cross-validation, the prediction accuracy of the models has been tested by using R^2, $RMSE$, MAE, and $MAPE$. In this study, for the first time in the literature, for a single dataset, maximum number of coal samples were utilized and GRNN and RBFNN methods were used in GCV prediction based on proximate analysis. The results showed that moisture and ash are the most discriminative predictors of GCV and the developed RBFNN-based models produce high performance for GCV prediction. Additionally, performances of the regression methods, from the best to the worst, were RBFNN, GRNN, MLP, and MLR.

Key words: Coal gross calorific value, regression, multiple linear regression, multilayer perceptron, general regression neural network, radial basis function neural network

1. Introduction

Coal is a lightweight, combustible, black or dark brown organic-origin rock consisting mainly of carbonized plant matter and found mainly in underground deposits with ash-forming minerals [1]. Energy demand of the world is increasing with every passing year. At the present time, the energy demand of the whole world is mostly met by fossil-based fuels such as fuel-oil, natural gas, and coal [2]. Due to its carbon content, coal is one of the most commonly used fossil fuel among other energy-supplier materials. It occupies the first place both in abundance and life cycle, and is considered the most important energy source in the long term. The demand for coal has increased considerably due to efforts to supply new coal-burning thermal electric power plants [1, 3]. Therefore, coal is an important and prevalent fuel in the world, which supplies about 40–45% of the planet's energy needs [1, 4]. The reasons why coal is mostly used in energy generation are its abundance and financial advantages and that coal is expected to remain the dominant energy source for the near future [5, 6]. Hence, the outstanding usage area of coal is the thermal power plants to generate electricity. However, depending on its rank (coal

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quality), coal can be used in different industries for different purposes such as cement making, coke production for metallurgical furnaces, or domestic heating. The usability of coal in different industries can be determined after coal analyses.

Characteristics of coal can be determined following the procedures in internationally acceptable test standards. The proximate and ultimate analyses are two kinds of test sets to determine the quality of coal. The moisture (M), ash (A), volatile matter (VM), fixed carbon (FC), and calorific values are measured in proximate analyses. On the other hand, carbon, hydrogen, nitrogen, sulphur, and oxygen contents are measured in ultimate analyses [2, 7]. However, the most important parameter among them is the calorific value of coal.

Calorific value is the heat capacity of a unit weight of coal after burning completely [7, 8]. It is usually referred to as gross calorific value (GCV) or higher heating value (HHV). Bomb calorimeter is used to measure the GCV of a coal sample. The standard Bomb calorimeter test method requires an expensive advanced test device and an experienced technician. On the other hand, M, A, VM, and FC can be obtained easily by using a simple laboratory oven and muffle furnace. These devices are easier and cheaper than a Bomb calorimeter and can be used by a laboratory technician. Similarly, very expensive instrumental analyzers and highly experienced technicians are required to carry out the ultimate analysis of a coal sample [9].

Since coal is a combustible organic-origin rock, it consists of organic and inorganic components together. Coal quality is a function of these organic and inorganic components and naturally there should be a correlation between them. For instance, if the inorganic components increase in a coal sample, the organic components decrease, specifically, if the ash content of a coal sample increases, the carbon content decreases and the calorific value decreases, and vice versa. This is of course not only correlation; there may be other relationships among coal properties. Therefore, many researches have been carried out to investigate these relationships by scholars. The relationships among coal properties have been studied by researchers and many equations and prediction models have been proposed to estimate the GCV of coal samples based on proximate analyses and/or ultimate analyses [2, 6, 7, 9–24].

The present study had two main purposes; the first one was to develop prediction models for GCV of coal using as-received basis proximate analysis results and the second one was to reveal the discriminative predictors of GCV. For this reason, multiple linear regression (MLR) and artificial neural networks (ANNs) (multilayer perceptron (MLP), general regression neural network (GRNN), and radial basis function neural network (RBFNN)) methods were applied to the developed models created by different combinations of the proximate analysis variables. In order to compare the results of the prediction models, multiple correlation coefficient (R^2), root mean square error ($RMSE$), mean absolute error (MAE), and mean absolute percentage error ($MAPE$) were utilized as performance metrics. Nevertheless, for a single dataset, the maximum number of coal samples were utilized in this study when compared to previous works in the literature. In addition, in this study, GRNN and RBFNN methods were first used in GCV prediction based on proximate analysis results.

2. Dataset generation

The dataset used in this study was derived from the coal database (COALQUAL Version 3.0) which was developed by U.S. Geological Survey (USGS) Energy Resources Program. The COALQUAL database consists of analysis results obtained from different ranks of coal including lignite, subbituminous, bituminous, anthracite coals and includes the proximate and ultimate analysis results, as well as GCV on as-received basis. All analyses were conducted in commercial testing laboratories in accordance with the ASTM standards. The database is located at the official web site, http://ncrdspublic.er.usgs.gov/coalqual/.

The samples where validation rating of proximate analysis and/or ultimate analysis was "Suspect" or "Incomplete Data" were excluded from the database. In this study, proximate analysis results were used to predict GCV of coal. The dataset was composed by using 6520 proximate analyses results of samples including variables M, A, VM, FC, and GCV.

For this dataset, M, A, VM, and FC were selected as predictor variables and GCV was selected as the target variable. Although the predictor variables M, A, and VM were obtained by measuring in laboratory environment, the predictor variable FC was calculated using M, A, and VM. The calculation of FC is given in Eq. (1).

$$FC = 100 - (M + A + VM). \tag{1}$$

Table 1 presents the descriptive statistics of the dataset. The scatter plots of GCV vs. predictor variables are shown in Figure 2.

Table 1. Descriptive statistics of the dataset.

Category	Variable name	Minimum value	Maximum value	Range	Mean	Standard deviation
Predictor variables	M (%)	0.40	52.50	52.10	8.13	9.97
	A (%)	0.90	54.70	53.80	11.80	7.30
	VM (%)	3.00	51.96	48.96	32.02	6.51
	FC (%)	12.70	87.00	74.30	48.06	11.26
Target variable	GCV (Mj/kg)	8.82	36.26	27.44	26.91	5.35

3. Methodology and prediction models

3.1. Methodology

For the analyzed dataset, by using combinations of the predictor variables M, A, VM, and FC, 11 models were created to determine the effectiveness of each predictor variable for predicting GCV of coal. Table 2 shows the prediction models for GCV and the predictor variables that each model has.

In this study, MLR-, MLP-, GRNN-, and RBFNN-based prediction models were utilized for predicting GCV of coal. The results of the presented GCV prediction models were compared with each other.

10-fold cross-validation was used for satisfying the generalization of GCV prediction models. The prediction performance of the presented GCV models were computed by using the following metrics: R^2, $RMSE$, MAE, and $MAPE$. The formulas of these performance metrics are shown in Eqs. (2)–(5).

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(Y_i - Y_i')^2}{\sum_{i=1}^{n}(Y_i - \bar{Y})^2}, \tag{2}$$

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(Y_i - Y_i')^2}, \tag{3}$$

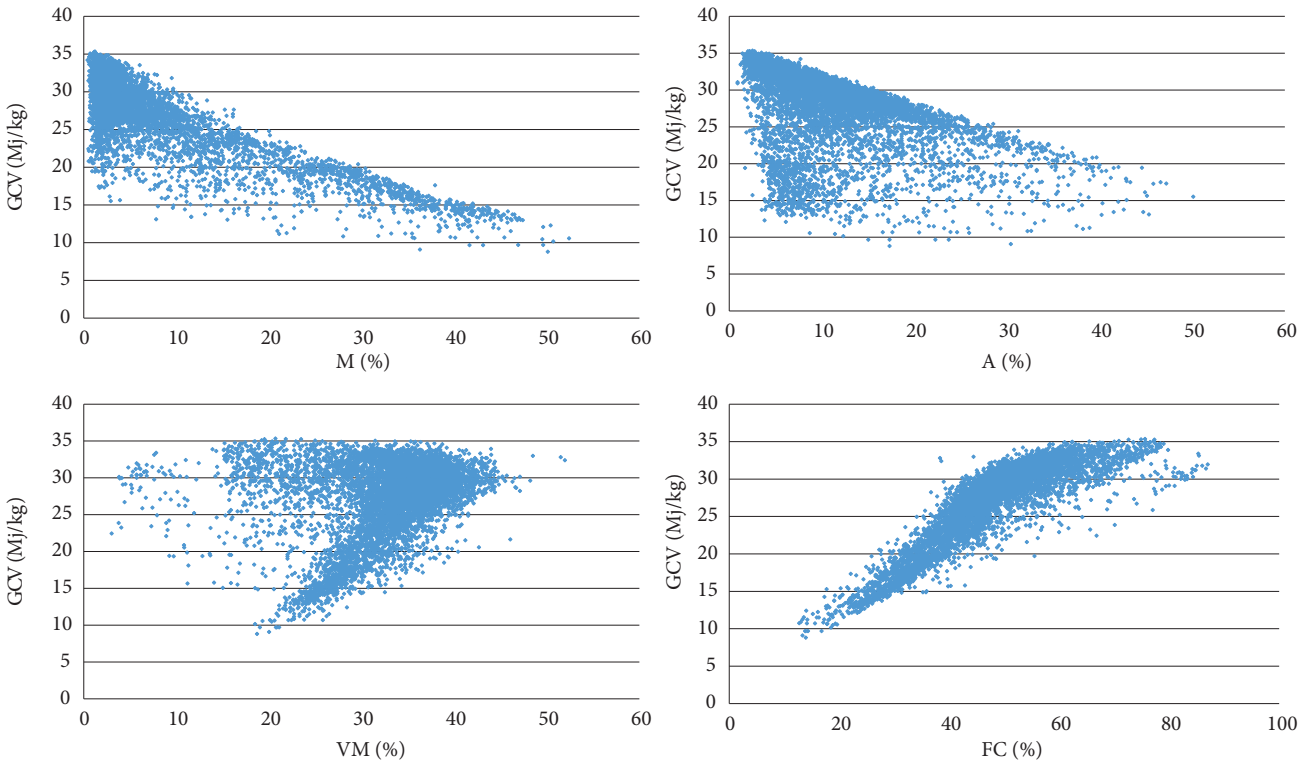

Figure. The scatter plots of GCV vs. predictor variables for dataset.

$$MAE = \frac{1}{n}\sum_{i=1}^{n}|Y_i - Y_i'|, \tag{4}$$

$$MAPE = \frac{1}{n}\sum_{i=1}^{n}\frac{|Y_i - Y_i'|}{Y_i}. \tag{5}$$

In Eqs. (2)–(5), the measured GCV value is represented with Y, the predicted GCV value is represented by Y', the mean of the measured values of GCV is represented by $\bar{Y}$, and the number of instances in a test set is given by n.

As stated before, 10-fold cross-validation was applied to the original dataset. The dataset was randomly split into 10 subsets. The training data was formed using 9 subsets, while the tenth subset was used as the testing data. This procedure was carried out exactly 10 times so that the testing data included every 10 subsets. The final value of the performance measures of each prediction model was obtained by averaging the results of each fold.

A predictive modeling and forecasting software, DTREG, was used to perform all the experiments presented in this study. DTREG is a robust application that is capable of building a variety of machine learning methods [25].

3.2. MLR-based models for predicting GCV

Due to its flexibility and power, MLR is frequently used in statistical analysis. It is an extension of the simple linear regression model in that it uses two or more dependent variables in a prediction formula to estimate

Table 2. Overview of the GCV prediction models along with the predictor variables for the dataset.

Models	Predictor variables
Model 1	M, A, VM, FC
Model 2	M, A, VM
Model 3	M, A, FC
Model 4	M, VM, FC
Model 5	A, VM, FC
Model 6	M, A
Model 7	M, VM
Model 8	M, FC
Model 9	A, VM
Model 10	A, FC
Model 11	VM, FC

a desired variable. Even with complicated regression models that include a large number of variables, MLR requires little effort to generate predictions.

3.3. ANN-based models for predicting GCV

ANNs constitute a class of flexible nonlinear models designed to mimic biological neural systems [26]. They have been widely used and applied to resolve many different problems in different areas. There are many types of ANNs for modeling function approximation of the problems [27]. In this context, three types of neural networks, namely MLP, GRNN, and RBFNN, were used to confirm the usefulness of the proposed prediction models in this study.

3.3.1. MLP-based models for predicting GCV

The most common ANN model is the MLP network training with back-propagation algorithm. It is a flexible and general-purpose type of ANN composed of one input layer, one or more hidden layers, and one output layer [28]. In general, an MLP will not achieve optimal results if the number of neurons in every hidden layer is not adjusted properly. To build an accurate MLP model, it is very important to select the optimal number of neurons in the hidden layer. There is no rule for determining the number of neurons in the hidden layer; it varies according to the complexity of the problem [29]. For this study, logistic and linear functions were respectively selected as activation functions in the hidden and output layers. Additionally, the optimal number of neurons in the hidden layer was chosen by trial-and-error for each model and found by comparing the performances of different networks that use values of number of neurons shown in Table 3.

3.3.2. GRNN-based models for predicting GCV

A memory-based network, GRNN provides predictions of continuous variables and converges to the underlying regression surface. It is established based on a one-pass learning algorithm with a highly parallel structure and does not require back-propagation procedure to learn error of the training data. The GRNN uses an algorithm that provides smooth transitions from one observed value to another, even with sparse data in a

multidimensional measurement space, and it can be applied for any regression problem in which an assumption of linearity is not justified [30].

Each GRNN consists of four layers which are input layer, pattern layer, summation layer, and output layer. The input and pattern layers are fully connected and each node on the pattern layer represents a radial basis function (RBF) (also called the kernel function) [31]. Although different RBF types are available, the most common kernel function is the Gaussian function. The width of the Gaussian function is a parameter controlling the smoothness properties of the function and is called the smoothing factor (σ) [31].

For this study, the optimal smoothing factor for each node in the pattern layer was determined by using an iterative procedure for each model and found by comparing the performances of different networks that use values of the smoothing factor shown in Table 3.

3.3.3. RBFNN-based models for predicting GCV

An RBFNN is a feed-forward neural network that consists of three layers which are input layer, hidden layer, and output layer. An RBFNN is similar to a special case of multilayer feed-forward neural networks, but different in terms of node characteristics and learning algorithm [32]. The neurons in the hidden layer contain Gaussian functions whose outputs are inversely proportional to the distance from the center of the neuron. RBFNNs are very similar to GRNNs. The main difference is that GRNN has one neuron for each point in the training file, whereas RBFNN has a variable number of neurons that is usually much less than the number of training points [25].

Four different parameters are determined in the training phase of the RBFNN model. These parameters are the number of neurons in the hidden layer, the coordinates of the center of each hidden-layer RBF function, the radius (spread) of each RBF function in each dimension, and the weights applied to the RBF function outputs as they are passed to the output layer [25].

For this study, the optimal parameters for each RBFNN-based model were determined by trial-and-error and found by comparing the performances of different networks that use parameters shown in Table 3. The details of methodology of finding the optimal parameters for RBFNN can be found in user manual [25].

Table 3. Values/ranges of the parameters for the ANN-based prediction models.

Type of ANN	Parameter description	Value/Range
MLP	Number of neurons in the hidden layer	[3, 15]
	Activation function for hidden layer	Logistic
	Activation function for output layer	Linear
	Training method	Scaled conjugate gradient
GRNN	Kernel function	Gaussian
	Smoothing factor (σ)	[0.0001, 10]
RBFNN	Kernel function	Gaussian
	Maximum number of neurons in the hidden layer	100
	The radius (spread) of each RBF in each dimension	[0.01, 400]
	Regularization parameter (Lambda)	[0.001, 10]

4. Results and discussion

4.1. Results

To evaluate the effectiveness of the presented GCV prediction models based on combinations of proximate analysis variables, experiments were conducted on the dataset by using MLR, MLP, GRNN, and RBFNN methods. For comparison purposes, Tables 4–7 illustrate the performance measures (R^2, $RMSE$, MAE, and $MAPE$) values for the prediction models employed by regression methods.

Table 4. Performance measures for GCV prediction models using MLR on the dataset.

Models	Predictor variables	R^2	$RMSE$ (Mj/kg)	MAE (Mj/kg)	$MAPE$ (%)
Model 1	M, A, VM, FC	0.973	0.86	0.62	2.65
Model 2	M, A, VM	0.973	0.86	0.62	2.65
Model 3	M, A, FC	0.973	0.86	0.62	2.65
Model 4	M, VM, FC	0.973	0.86	0.62	2.65
Model 5	A, VM, FC	0.973	0.86	0.62	2.64
Model 6	M, A	0.965	0.97	0.75	3.09
Model 7	M, VM	0.706	2.85	2.16	8.88
Model 8	M, FC	0.861	1.96	1.48	5.83
Model 9	A, VM	0.196	4.72	3.64	16.38
Model 10	A, FC	0.766	2.54	2.04	8.33
Model 11	VM, FC	0.964	0.98	0.74	3.09

Table 5. Performance measures for GCV prediction models using MLP on the dataset.

Models	Predictor variables	R^2	$RMSE$ (Mj/kg)	MAE (Mj/kg)	$MAPE$ (%)
Model 1	M, A, VM, FC	0.984	0.65	0.47	1.89
Model 2	M, A, VM	0.984	0.65	0.47	1.90
Model 3	M, A, FC	0.984	0.65	0.47	1.91
Model 4	M, VM, FC	0.984	0.64	0.46	1.89
Model 5	A, VM, FC	0.984	0.65	0.47	1.89
Model 6	M, A	0.980	0.74	0.56	2.20
Model 7	M, VM	0.735	2.70	2.04	8.07
Model 8	M, FC	0.902	1.64	1.22	4.73
Model 9	A, VM	0.263	4.51	3.53	15.66
Model 10	A, FC	0.910	1.57	1.09	4.43
Model 11	VM, FC	0.969	0.92	0.67	2.75

4.2. Discussion

In studies [6, 9, 16, 17, 20, 33] in this area, often only one performance metric was used to demonstrate the accuracy of the model for GCV prediction. One of them, R^2, is the most commonly used performance metric,

Table 6. Performance measures for GCV prediction models using GRNN on the dataset.

Models	Predictor variables	R^2	$RMSE$ (Mj/kg)	MAE (Mj/kg)	$MAPE$ (%)
Model 1	M, A, VM, FC	0.984	0.64	0.46	1.86
Model 2	M, A, VM	0.984	0.65	0.47	1.87
Model 3	M, A, FC	0.984	0.64	0.46	1.86
Model 4	M, VM, FC	0.984	0.65	0.46	1.87
Model 5	A, VM, FC	0.982	0.70	0.49	1.97
Model 6	M, A	0.979	0.75	0.56	2.21
Model 7	M, VM	0.744	2.66	1.98	7.82
Model 8	M, FC	0.909	1.58	1.16	4.50
Model 9	A, VM	0.358	4.21	3.18	14.03
Model 10	A, FC	0.914	1.54	1.06	4.32
Model 11	VM, FC	0.969	0.91	0.66	2.69

Table 7. Performance measures for GCV prediction models using RBFNN on the dataset.

Models	Predictor variables	R^2	$RMSE$ (Mj/kg)	MAE (Mj/kg)	$MAPE$ (%)
Model 1	M, A, VM, FC	0.986	0.62	0.44	1.76
Model 2	M, A, VM	0.986	0.62	0.44	1.76
Model 3	M, A, FC	0.986	0.62	0.44	1.76
Model 4	M, VM, FC	0.986	0.62	0.44	1.76
Model 5	A, VM, FC	0.985	0.61	0.44	1.79
Model 6	M, A	0.981	0.72	0.54	2.11
Model 7	M, VM	0.742	2.67	1.99	7.85
Model 8	M, FC	0.911	1.57	1.15	4.45
Model 9	A, VM	0.342	4.27	3.23	14.21
Model 10	A, FC	0.913	1.54	1.07	4.33
Model 11	VM, FC	0.970	0.90	0.66	2.68

but in general, it alone is not enough to interpret and compare prediction errors of models. As stated before in Section 3.1, four different performance metrics were utilized in this study. These metrics were used to compare the performances of the models developed in this study and to make comparisons with the performances of the models developed in other studies. The correlation coefficient and errors of the GCV prediction models were computed using R^2 and the triple of $RMSE$, MAE, $MAPE$, respectively. As the performance of the model increases, the correlation coefficient increases (converges to 1.0) and the error decreases (converges to 0.0).

As shown in Tables 4–7, all ANN-based prediction models are more accurate than the MLR-based models. According to these findings, it can be said that the prediction of GCV of coal has a nonlinear characteristic. In ANN-based prediction models, the RBFNN-based prediction model performs better than the MLP- and GRNN-based prediction models. The ranking in the performances of the regression methods for GCV prediction, from the best to the worst, is as follows: RBFNN, GRNN, MLP, and MLR.

The errors of the models including quadruple and triple combinations of the predictor variables (Model 1 to Model 5) are almost similar and these models have higher correlations and lower errors than the ones including dual combinations of predictor variables.

In general, it is desirable that the values of all the predictor variables used for a prediction model are collected by measuring. However, sometimes it is seen that the value of a predictor variable is obtained by calculation. If the calculation of a predictor variable value depends only on the other predictor variables, the inclusion of that variable to the model does not cause a noticeable increase in the accuracy of the prediction model. According to this approach, when comparing the performances of Model 1 and Model 2 for all regression methods, it is clear that the FC variable has a negligible effect on prediction. In addition, since the FC is derived from the other predictor variables (M, A, and VM), it gives higher correlations and lower errors in the other models including the FC variable.

The results show that addition of the predictor variables M and A into the prediction models has a strong positive effect and distinctively reduces error for GCV prediction. The ranking in the performance of the predictor variables except FC for GCV prediction, from the highest to the lowest, is as follows: M, A, and VM.

4.3. Comparing the results

It is to be noted that for most of the studies suggested in the literature, it is not possible to compare their prediction results directly and in detail with those obtained in this study because each study *a*) uses different datasets which contain different numbers of samples, *b*) uses different datasets which have different data characteristics (e.g., coal rank and region extracted), *c*) uses different datasets which have different predictor variables based on the analysis type, *d*) uses different types of prediction methods, and *e*) uses different data validation methods.

However, there are a few studies in the literature [6, 16, 20] that utilized the database COALQUAL Version 2.0 with 4540 entries which was a subset of the dataset used in this study (COALQUAL Version 3.0 with 6520 entries). In addition, these studies used the same predictor variables based on proximate analysis, whereas they utilized different prediction and validation methods. Hence, a comparison can be made between the performances of prediction models in this study and those of the mentioned studies [6, 16, 20].

Mesroghli et al. [16] developed a prediction model that includes the predictor variables M, A, and VM. Multivariable regression (corresponds to MLR) and feed-forward ANN (corresponds to MLP) methods were applied to this model. In contrast to the present study, they showed that the MLR-based model performs better than the MLP-based model for predicting GCV. For the MLR- and MLP-based models, the presented R^2 values were respectively 0.97 and 0.95 in their study, whereas the presented R^2 values were respectively 0.973 and 0.984 in the present study. According to these results, performances of the MLR-based models were similar to each other; however, prediction results of the MLP-based model developed in the present study are more accurate than those of their study.

Tan et al. [6] proposed a prediction model based on support vector regression (SVR). According to their study, for the SVR-based prediction model that includes the predictor variables M, A, VM, and FC, the average absolute error percentage (corresponds to $MAPE$) was 2.42%. In the present study, for Model 1 given in Table 7, $MAPE$ value was 1.76%. In another study [20], Matin and Chelgani developed prediction models by using the multivariable regression (corresponds to MLR) and random forest (RF) methods. They also used M, A,

and VM as predictor variables. The reported values of R^2 for the MLR- and RF-based models were the same (0.97). According to the present study, for the RBFNN-based prediction model (Model 2 given in Table 7), the R^2 value was obtained as 0.986. Hence, the results showed that the RBFNN-based prediction model proposed in the present study performs better than the SVR-based prediction model proposed in [6] and the RF-based prediction model proposed in [20].

5. Conclusion

The objective of this work was to develop new models using MLR, MLP, GRNN, and RBFNN for prediction and show the discriminative predictor variables of GCV. The dataset was generated by using proximate analyses results and GCV, belonging to 6520 coal samples in as-received basis. By using the combinations of the predictor variables (M, A, VM, and FC), 11 models were developed for predicting GCV. For comparison purposes, the aforementioned regression methods were implemented to the models. 10-fold cross-validation was utilized for satisfying the generalization capability of the developed models. The performances of the prediction models were stated by calculating the values of metrics R^2, $RMSE$, MAE, and $MAPE$.

Among the regression models, prediction models based on RBFNN exhibited better performances than the models developed by using MLR, MLP, or GRNN, irrespective of which variant of the predictor variable was used. The MLR-based prediction models exhibited the worst performances for predicting GCV. On the other hand, MLR produced faster results compared to the other regression methods for prediction.

For all the results regarding regression methods, the same comments can be made for the effect of the relevant predictor variables, regardless of the regression method used. So the results reveal that, among the predictor variables, M and A have more positive effect on the performance of GCV prediction. Besides, as discussed before, the variable FC has negligible or strong effect (since FC has information about other predictor variables) in different models. The ranking in the performance of the predictor variables except FC for GCV prediction, in descending order, is M, A, and VM.

Taking into account the performances of all the models proposed in this study, the RBFNN-based predictor model that includes the predictor variables M, A, and VM can be the most suitable model for predicting GCV of coal.

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