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Research Article

Multivariable Regression and Adaptive Neurofuzzy Inference System Predictions of Ash Fusion Temperatures Using Ash Chemical Composition of US Coals

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In this study, the effects of ratios of dolomite, base/acid, silica, SiO_2/Al_2O_3, and Fe_2O_3/CaO, base and acid oxides, and 11 oxides (SiO_2, Al_2O_3, CaO, MgO, MnO, Na_2O, K_2O, Fe_2O_3, TiO_2, P_2O_5, and SO_3) on ash fusion temperatures for 1040 US coal samples from 12 states were evaluated using regression and adaptive neurofuzzy inference system (ANFIS) methods. Different combinations of independent variables were examined to predict ash fusion temperatures in the multivariable procedure. The combination of the "11 oxides + (Base/Acid) + Silica ratio" was the best predictor. Correlation coefficients (R^2) of 0.891, 0.917, and 0.94 were achieved using nonlinear equations for the prediction of initial deformation temperature (IDT), softening temperature (ST), and fluid temperature (FT), respectively. The mentioned "best predictor" was used as input to the ANFIS system as well, and the correlation coefficients (R^2) of the prediction were enhanced to 0.97, 0.98, and 0.99 for IDT, ST, and FT, respectively. The prediction precision that was achieved in this work exceeded that reported in previously published works.

1. Introduction

Usually, coal quality for coal used for the generation of electricity refers to differences in heating value, grindability, sulfur content, and ash fusion characteristics. The potential for slagging on furnace walls is related to the ash composition and temperature. Ash fusibility is a factor that can be used to determine the performance of coals related to slagging [1].

ASTM Standard D1857, which specifies the experimental method for determining ash fusion temperatures (AFTs), is based on the gradual thermal deformation of a pyramid-shaped ash sample in either an oxidizing or reducing atmosphere. The test results can be reported for four temperatures: (1) the initial deformation temperature (IDT), which is the temperature at which the first rounding of the apex of the cone occurs and the pyramid begins to demonstrate evidence of deformation; (2) the softening temperature (ST) or fusion temperature (FT), which is the temperature at which the cone has fused and the height equals the width; (3) the hemispherical temperature (HT), which is the temperature at which the cone has fused into a hemispherical lump and the height is equal to half of the width at the base; and (4) the fluid temperature (FT), which is the temperature at which the fused mass has spread out in a nearly flat layer [2].

The temperature difference between the initial deformation temperature and the fluid temperature gives information on the type of deposit to be expected on the surfaces of the furnace tubes [3]. A small temperature difference indicates a thin and difficult-to-remove slag, whereas greater temperature differences indicate less adhesive deposits. The softening temperature is a criterion used to indicate the ease with which deposits can be removed from heat-transfer surfaces. If ash particles arrive at heat-absorbing surfaces at temperatures below their softening temperature, ash removal is relatively easy. If the ash particles arrive at these surfaces at temperatures above their softening temperature, the resulting deposit is more difficult to remove [3].

Although AFTs can be used to predict the slagging and fouling properties of coals, the reproduction of AFTs for

a given coal in different laboratories may differ by 100 ± 20°C [4–6].

Many researchers have tried to predict ash fusion temperatures based on the chemical composition of the coal [4, 7–11]. If reliable, correlative results could be established, and they could serve as a guide for coal combustion operation and eliminate the need for duplicate laboratory tests. Winegartner and Rhodes (1975) used regression analysis in an attempt to correlate ash fusibility with its chemical composition for a large number of ash samples from coal in United States (eastern and western coal), but their correlations were poor when they were applied to other coal ash samples from different sources [4]. Gray (1987) predicted ash fusion temperatures from the ash compositions of some New Zealand coals using three techniques of ternary equilibrium phase diagrams, stepwise regression analyses, and multiple regression analyses; these techniques were capable of predicting ash fusion temperatures to within 40°K [7]. Seggiani (1999) used a database that included about 300 ash samples from coals of different sources and biomasses. The proposed correlations predicted ash fusion temperatures and the temperatures of critical viscosity with a correlation (R) of 0.8. It was also mentioned that a small number of samples with similar compositions can give good results by the statistical approach, but the application to a large number of samples from different sources and with a wide range of compositions can decrease the effectiveness of the method [8].

Lolja et al. (2002) analyzed the ash fusion temperatures for 17 Albanian coals using oxides analysis from various perspectives, such as discrete species, acids and bases, crystal components, fluxing agents, and cement constituents, and in accordance with the periodic table. It was shown that deformation temperature and flow temperature correlated well with basic and acidic oxides, respectively. Also, AFTs decreased as basic oxide content increased [9].

G. Özbayoğlu and M. E. Özbayoğlu (2006) used the chemical composition of ash (eight oxides) and coal parameters, such as ash content, specific gravity, Hardgrove index, and mineral matter content, to predict AFTs and showed that nonlinear correlations are better than linear correlations for estimating ash fusion temperatures [11].

Liu et al. (2007) used the Ant Colony Optimization-Back Propagation (ACO-BP) neural network approach to model coal ash fusion temperatures based on the chemical compositions of the ashes. Data on 80 typical Chinese coal ash samples were used for training and testing. The results showed that the ACO-BP neural network can obtain better performance than empirical formulas and BP neural networks [12].

The aim of this research is to provide new correlations for the prediction of AFTs by using an extensive coal ash composition database. The specific questions we sought to answer in this research are listed below.

(i) Is it possible to predict AFTs that have acceptable correlations using an extensive database that consists of data from the chemical analyses of 1040 coal ash samples from 12 states in the USA?

(ii) Which variables have significant positive and negative effects?

(iii) Which combination of predictive indices is the best for predicting AFTs for a large number of samples from different sources?

(v) Is it possible to achieve better correlations and decrease errors using adaptive neurofuzzy inference system (ANFIS) modeling?

2. Experimental

The database that was used to test the proposed approaches was obtained from the U.S. Geological Survey Coal Quality (COALQUAL) project of the USGS. A total of 1040 sets of coal ash analyses were used. The database, including the major and minor constituents from the oxide analyses, that is, SiO_2, Al_2O_3, CaO, MgO, MnO, Na_2O, K_2O, Fe_2O_3, TiO_2, P_2O_5, and SO_3, as well as ash fusion temperatures, is also available in the electronic Appendix to this paper.

The sampling procedures and the chemical analytical methods used can be found at the following web address: http://energy.er.usgs.gov/products/databases/CoalQual/index.htm. The number of samples and the range of AFTs for different states are shown in Table 1.

3. Methods

3.1. Input Variables. Table 2 shows the range of input variables, which consists of 11 oxides, base, and acid, as well as base/acid, dolomite, silica, SiO_2/Al_2O_3, and Fe_2O_3/CaO ratios, that was used to predict AFTs; these are the variables that were commonly used in previously published works [4, 7–12]. Each variable was examined separately and in combination with the other variables. The input variables in separate forms are as follows:

$$\text{Oxide} = SiO_2 + Al_2O_3 + CaO + MgO + MnO + Na_2O + K_2O + Fe_2O_3 + TiO_2 + P_2O_5 + SO_3 \quad (1)$$

$$\text{Base} = Fe_2O_3 + CaO + MgO + K_2O + Na_2O + MnO \quad (2)$$

$$\text{Acid} = SiO_2 + Al_2O_3 + TiO_2 + P_2O_5 + SO_3 \quad (3)$$

$$\frac{\text{Base}}{\text{Acid}} = \frac{(Fe_2O_3 + CaO + MgO + K_2O + Na_2O + MnO)}{(SiO_2 + Al_2O_3 + TiO_2 + P_2O_5 + SO_3)} \quad (4)$$

$$\text{Dolomite ratio (Dr)} = \frac{(CaO + MgO)}{(Fe_2O_3 + CaO + MgO + K_2O + Na_2O)} \quad (5)$$

$$\text{Silica ratio (Sr)} = \frac{SiO_2}{(SiO_2 + Fe_2O_3 + CaO + MgO)} \quad (6)$$

Table 1: Number of samples and range of AFTs in different US states.

State	IDT (°F)		ST (°F)		FT (°F)		Number of samples
	Min	Max	Min	Max	Min	Max	
Alabama	1790	2910	1930	2910	2020	2910	519
Alaska	1966	2800	2075	2800	2160	2800	40
Arizona	2130	2290	2160	2400	2190	2480	5
Arkansas	2070	2660	2080	2700	2120	2730	21
Colorado	1960	2910	2050	2910	2230	2910	134
Georgia	2070	2800	2190	2800	2280	2800	24
Illinois	1951	2276	2024	2353	2072	2425	5
Indiana	1955	2680	1980	2785	2000	2800	33
Iowa	1905	2265	1970	2375	2020	2490	27
Kansas	1855	2050	1960	2100	2050	2170	10
Kentucky	1820	2910	1940	2800	2080	2848	204
Maryland	1940	2790	2020	2800	2320	2800	18

Table 2: The range of variables in 1040 US coal ash analyses.

Variable	Min	Max	Mean	Standard deviation	Variance
SiO_2 (%)	13.00	80.00	42.63	12.25	150.07
Al_2O_3 (%)	1.60	46.00	23.73	6.97	48.59
CaO (%)	0.12	29.00	3.90	4.66	21.71
MgO (%)	0.12	7.90	1.08	0.81	0.66
MnO (%)	0.00	1.20	0.04	0.08	0.007
Na_2O (%)	0.02	6.80	0.54	0.70	0.49
K_2O (%)	0.01	6.80	1.78	0.98	0.96
Fe_2O_3 (%)	0.60	69.00	16.33	13.87	192.30
TiO_2 (%)	0.08	5.60	1.12	0.49	0.24
P_2O_5 (%)	0.00	8.00	0.48	0.68	0.47
SO_3 (%)	0.00	28.00	3.41	3.65	13.31
Base	2.23	70.98	23.68	14.51	210.54
Acids	23.26	98.61	71.38	15.9	252.81
Base/Acid	0.02	2.72	0.4166	0.405	0.164
Dolomite ratio	0.01	0.87	0.23	0.18	0.03
Silica ratio	0.17	0.99	0.67	0.20	0.04
SiO_2/Al_2O_3	0.84	23.75	1.90	0.91	0.84
Fe_2O_3/CaO	0.13	110.00	10.40	14.15	200.24
IDT	1790	2910	2368.58	282.73	79936.25
ST	1930	2910	2455.19	266.04	70777.28
FT	2000	2910	2543.83	229.20	52532.64

$$\frac{SiO_2}{Al_2O_3} \quad (7)$$

$$\frac{Fe_2O_3}{CaO} \quad (8)$$

3.2. Regression. Linear regression estimates the coefficients of the linear equation, involving one or more independent variables, to have a reliable prediction of the value of the dependent variable. All variables must pass the tolerance criterion to be entered in the equation, regardless of the entry method specified. The default tolerance level is 0.0001. Also, a variable is not entered if it would cause the tolerance of another variable already in the model to drop below the tolerance criterion. All independent variables selected are added to a single regression model. However, different entry methods can be specified for different subsets of variables. Method selection allows specifying how independent variables are entered into the analysis. Using different methods, a variety of regression models can be selected from the same set of variables [13].

Nonlinear regression is a method of finding a nonlinear model of the relationship between the dependent variable and a set of independent variables. Unlike traditional linear regression, which is restricted to estimating linear models, nonlinear regression can estimate models with arbitrary relationships between independent and dependent variables. This is accomplished using iterative estimation algorithms [13].

In this study, both linear and nonlinear regression analyses were used to develop correlations between the chemical compositions of various ashes and their fusion temperatures by using the stepwise regression in the SPSS software package. A stepwise procedure for selecting variables was used, and the variables were entered sequentially into the model. The first variable considered for use in the equation was the one with the largest positive or negative correlation with the dependent variable. This variable was entered into the equation only if it satisfied the criterion for entry. The next variable, with the largest partial correlation, was considered as the second input to the equation. The procedure stops when there are no variables that meet the entry criterion [13].

3.3. Adaptive Neurofuzzy Inference System (ANFIS). In the artificial intelligence field, the term "neurofuzzy" refers to combinations of artificial neural networks and fuzzy logic. Fuzzy modeling and neural networks have been recognized as powerful tools that can facilitate the effective development of models and integrate information from different sources, such as empirical models, physical laws, or measurements and heuristics [14]; these two tools were combined in order to achieve readability and learning ability at the same time [15].

The neurofuzzy in the fuzzy modeling research field is divided into two areas: linguistic fuzzy modeling that is focused on interpretability, mainly the Mamdani model, and precise fuzzy modeling that is focused on accuracy, mainly the Takagi-Sugeno-Kang (TSK) model [16].

ANFIS is an architecture that is functionally equivalent to a Takagi-Sugeno-Kang-type fuzzy rule base [17]; it is a class of adaptive, multilayer, feed-forward networks that is functionally equivalent to a fuzzy inference system.

A fuzzy rule in a Sugeno fuzzy model has the form of:

$$\text{if } x \text{ is } A \text{ and } y \text{ is } B, \text{ then } z = f(x, y),$$

where A and B are input fuzzy set in antecedent and, usually, $z = f(x, y)$ is a zero- or first-order polynomial function in the consequent. The fuzzy reasoning procedure for the first-order Sugeno fuzzy model and equivalent ANFIS structure is shown in Figure 1.

Here, the defuzzification procedure in the Mamdani fuzzy model is replaced by the operation of the weighted average in order to avoid the time-consuming procedure of defuzzification. Defuzzification refers to the way a crisp value is extracted from a fuzzy set as a representative value [17].

More details about the ANFIS architecture, learning algorithm, and training methods were described by [15, 17].

4. Results and Discussion

4.1. Relationships between AFTs and Individual Input Variables. By the least squares mathematical method, the correlation coefficients (R^2) of SiO_2, Al_2O_3, CaO, MgO, MnO, Na_2O, K_2O, Fe_2O_3, TiO_2, P_2O_5, SO_3, and base and acid oxides, as well as base/acid, dolomite, silica, SiO_2/Al_2O_3, and Fe_2O_3/CaO ratios for AFTs were determined. Table 3 shows the relationship between individual input variables and AFTs in both linear and nonlinear (quadratic and cubic) procedures. Exponential, logarithmic, logistic, inverse, compound, power, and growth nonlinear procedures were examined as well; the results were not better than the results obtained using quadratic and cubic procedures.

It can be seen that the correlations of SiO_2, Al_2O_3, Fe_2O_3, base, and acid, as well as base/acid and silica ratios, with AFTs are significant. The other parameters are not significant. Relationships of the above-mentioned significant variables with IDT, which is representative of AFTs, are shown in Figure 2. For the ST and FT temperatures, the effects of the variables were similar. According to Table 3 and Figure 2, it can be seen that the correlations between the AFTs and significant variables should be nonlinear in the prediction of AFTs. As a general result in IDT, ST, and FT predictions, the variables of acid oxide, silica ratio, Al_2O_3, and SiO_2 are the most significant variables, in that order, with positive effects on AFTs, and the variables of base oxide, base/acid, and Fe_2O_3 are the most significant variables, with negative effects on AFTs.

4.2. Multivariable Regression. The multivariable procedure was used to evaluate the possibility of achieving better prediction accuracy than the single-variable procedure. Different combinations of the most significant variables (Table 3 and Figure 2) were examined. Finally, the following eight combinations were determined as the suitable predictors:

$$\text{Input 1: 11 oxides: } SiO_2 + Al_2O_3 + CaO + MgO + MnO + Na_2O + K_2O + Fe_2O_3 + TiO_2 + P_2O_5 + SO_3 \quad (9)$$

$$\text{Input 2: Ln (Sr)} + \text{Dr} + \text{Ln}\left(\frac{1}{\text{Base}}\right) + \text{Acid} \quad (10)$$

$$\text{Input 3: Sr} + K_2O + Na_2O + SO_3 + MnO + Al_2O_3 + TiO_2 + P_2O_5 \quad (11)$$

$$\text{Input 4: Ln (Sr)} + SiO_2 + Al_2O_3 + Fe_2O_3 \quad (12)$$

$$\text{Input 5: Ln (Base)} + SiO_2 + Al_2O_3 + TiO_2 + P_2O_5 \quad (13)$$

$$\text{Input 6: Ln (Base)} + \text{Ln (Sr)} + SiO_2 + Al_2O_3 + Fe_2O_3 \quad (14)$$

$$\text{Input 7: Ln (Sr)} + \text{Ln (Acid)} + SiO_2 + Al_2O_3 + Fe_2O_3 \quad (15)$$

$$\text{Input 8: 11 oxide} + \left(\frac{\text{Base}}{\text{Acid}}\right) + \text{Sr} \quad (16)$$

Table 4 shows the correlation coefficient, standard deviation, mean, and residual (% difference between actual and predicted AFTs) less than 120°F for each of the inputs (see (9) to (16)) in both linear and nonlinear (quadratic) situations. The cubic equations did not improve the accuracy significantly.

The results show that input 8 (see (16)) was the best predictor based on the correlation coefficient and a residual of less than 120°F in the nonlinear condition. Equations (17)

Table 3: Correlation coefficients (R^2) between AFTs and independent variables.

Variable	IDT			ST			FT		
	Linear	Quadratic	Cubic	Linear	Quadratic	Cubic	Linear	Quadratic	Cubic
SiO_2	0.60	0.602	0.625	0.64	0.64	0.67	0.68	0.71	0.72
Al_2O_3	0.62	0.64	0.65	0.68	0.69	0.70	0.75	0.75	0.77
CaO	0.17	0.20	0.21	0.18	0.22	0.22	0.24	0.27	0.28
MgO	0.018	0.02	0.02	0.018	0.02	0.03	0.02	0.03	0.04
MnO	0.11	0.17	0.23	0.12	0.19	0.25	0.14	0.23	0.29
Na_2O	0.006	0.008	0.009	0.008	0.009	0.01	0.01	0.011	0.014
K_2O	0.02	0.025	0.046	0.035	0.036	0.06	0.06	0.06	0.08
Fe_2O_3	0.60	0.67	0.68	0.63	0.68	0.68	0.63	0.65	0.65
TiO_2	0.26	0.39	0.41	0.29	0.44	0.46	0.31	0.49	0.53
P_2O_5	0.001	0.004	0.007	0.001	0.006	0.008	0	0.004	0.005
SO_3	0.18	0.24	0.25	0.21	0.26	0.27	0.27	0.32	0.32
Base	0.76	0.84	0.84	0.80	0.86	0.86	0.84	0.85	0.85
Acid	0.74	0.81	0.81	0.79	0.84	0.84	0.84	0.85	0.85
Base/Acid	0.58	0.76	0.82	0.63	0.81	0.86	0.72	0.85	0.87
Dolomite ratio	0.015	0.11	0.21	0.01	0.11	0.21	0.003	0.07	0.18
Silica ratio	0.76	0.80	0.80	0.80	0.82	0.82	0.84	0.84	0.84
SiO_2/Al_2O_3	0.004	0.004	0.007	0.005	0.006	0.009	0.009	0.01	0.014
Fe_2O_3/CaO	0.09	0.10	0.10	0.09	0.10	0.10	0.07	0.07	0.07

Table 4: Precision of the prediction of AFTs using different inputs in the regression method.

		Parameter	Input 1	Input 2	Input 3	Input 4	Input 5	Input 6	Input 7	Input 8
IDT	L	R^2	0.820	0.827	0.835	0.837	0.856	0.866	0.868	0.87
		S.D.	119.39	117.65	114.78	114.27	107.421	103.49	102.77	101.93
		Mean	$3.25E-13$	$5.02E-13$	$-1.01E-12$	$-7.10E-13$	$5.27E-13$	$-1.54E-13$	$-1.28E-12$	−0.9647
		Re (%) < 120°F	65.38	70.19	68.94	72.40	75.38	78.46	77.98	76.82
	NL	R^2	0.887	0.839	0.883	0.88	0.865	0.889	0.89	0.891
		S.D.	95.395	113.606	96.264	97.997	103.911	94.256	93.836	93.272
		Mean	$-3.61E-6$	$1.97E-8$	$8.09E-7$	$-2.32E-9$	$-3.31E-8$	$-1.14E-7$	$-2.19E-8$	−0.11
		Re (%) < 120°F	81.73	72.30	80.86	80.28	77.59	81.44	82.11	83.26
ST	L	R^2	0.873	0.847	0.882	0.873	0.884	0.893	0.895	0.907
		S.D.	94.75	103.91	91.35	94.718	90.50	86.94	86.093	91.32
		Mean	$-8.40E-13$	$2.44E-13$	$-1.20E-12$	$-2.17E-13$	$-1.04E-12$	$1.68E-13$	$-1.27E-12$	−0.0742
		Re (%) < 120°F	81.05	75.56	82.69	80	81.82	85.19	84.90	87.69
	NL	R^2	0.914	0.862	0.909	0.903	0.874	0.916	0.917	0.917
		S.D.	78.109	98.792	79.87	82.706	100.233	77.138	76.744	76.792
		Mean	$4.59E-11$	$-2.78E-11$	$6.36E-7$	$-3.08E-8$	$-6.33E-9$	$1.37E-7$	$3.64E-8$	−0.08
		Re (%) < 120°F	88.84	77.21	88.75	85.38	78.17	88.46	89.32	89.51
FT	L	R^2	0.932	0.884	0.931	0.911	0.918	0.918	0.92	0.937
		S.D.	59.6	83.89	60.09	68.520	65.65	65.55	64.69	57.445
		Mean	$1.99E-13$	$6.02E-13$	$-7.01E-13$	$-3.33E-13$	$1.96E-13$	$-3.67E-13$	$-6.68E-13$	−0.0532
		Re (%) < 120°F	98.75	84.31	97.59	94.13	95.19	95.09	95.19	98.07
	NL	R^2	0.94	0.889	0.934	0.922	0.876	0.934	0.936	0.94
		S.D.	56.348	79.086	58.95	64	95.549	58.944	57.911	56.118
		Mean	$2.28E-10$	$2.27E-10$	$-1.17E-8$	$-1.53E-9$	$-1.75E-9$	$-2.59E-8$	$-1.20E-8$	−0.05
		Re (%) < 120°F	98.55	86.05	97.30	95	78.36	97.69	97.98	98.46

S.D.: Standard deviation.
L: Linear.
NL: Nonlinear.
Re (%) < 120°F: Residual (%, difference between actual and predicted AFTs) less than 120°F.

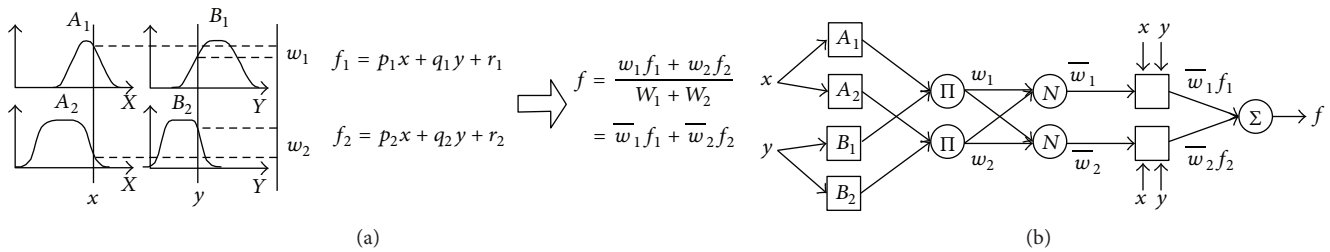

Figure 1: (a) The Sugeno fuzzy model reasoning and (b) equivalent ANFIS structure [17].

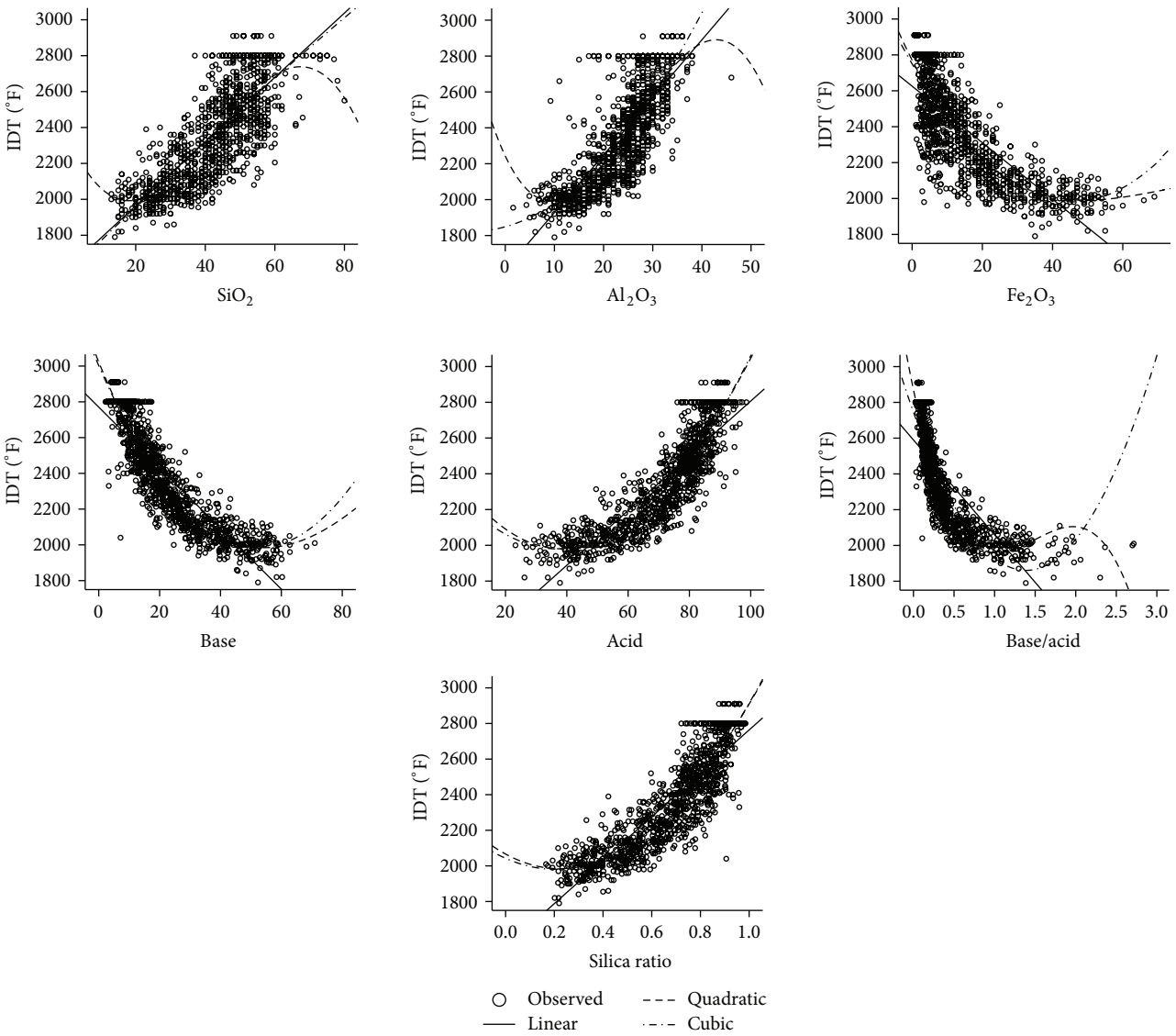

Figure 2: Relationship between IDT and significant independent variables.

Table 5: Comparison of ANFIS and nonlinear regression prediction of AFTs.

	IDT			ST			FT		
	R^2	S.D.	Re < 120°F (%)	R^2	S.D.	Re < 120°F (%)	R^2	S.D.	Re < 120°F (%)
Nonlinear regression	0.89	93.27	83.26	0.917	76.792	89.51	0.94	56.118	98.46
ANFIS	0.972	45.234	98.84	0.98	36.92	100	0.99	23.236	100

to (19) show the predictions of AFTs in nonlinear forms using input 8 in a multivariable, stepwise, regression procedure:

$$\begin{aligned}\text{IDT} = {} & 2011.604 - 9.474\,(\text{SiO}_2) + 9.128\,(\text{Al}_2\text{O}_3) \\ & - 8.777\,(\text{CaO}) - 44.451\,(\text{Na}_2\text{O}) - 9.105\,(\text{K}_2\text{O}) \\ & - 12.931\,(\text{Fe}_2\text{O}_3) - 16.727\,(\text{TiO}_2) - 2.452\,(\text{SO}_3) \\ & + 474.207\left(\frac{\text{Base}}{\text{Acid}}\right) - 61.729\,(\text{Sr}) + 0.133(\text{SiO}_2)^2 \\ & + 0.22(\text{Al}_2\text{O}_3)^2 + 0.198(\text{CaO})^2 - 1.256(\text{Na}_2\text{O})^2 \\ & - 5.796(\text{K}_2\text{O})^2 + 0.091(\text{Fe}_2\text{O}_3)^2 - 0.362(\text{TiO}_2)^2 \\ & + 0.225(\text{SO}_3)^2 - 120.481\left(\frac{\text{Base}}{\text{Acid}}\right)^2 + 633.978(\text{Sr})^2 \\ & R^2 = 0.891,\end{aligned} \tag{17}$$

$$\begin{aligned}\text{ST} = {} & 1951.611 - 5.859\,(\text{SiO}_2) + 13.322\,(\text{Al}_2\text{O}_3) \\ & - 9.197\,(\text{CaO}) - 53.724\,(\text{Na}_2\text{O}) - 13.494\,(\text{K}_2\text{O}) \\ & - 11.595\,(\text{Fe}_2\text{O}_3) - 5.207\,(\text{TiO}_2) - 1.026\,(\text{SO}_3) \\ & + 452.743\left(\frac{\text{Base}}{\text{Acid}}\right) + 183.388\,(\text{Sr}) + 0.111(\text{SiO}_2)^2 \\ & - 0.134(\text{Al}_2\text{O}_3)^2 + 0.150(\text{CaO})^2 - 0.728(\text{Na}_2\text{O})^2 \\ & - 4.305(\text{K}_2\text{O})^2 + 0.051(\text{Fe}_2\text{O}_3)^2 - 2.414(\text{TiO}_2)^2 \\ & + 0.732(\text{SO}_3)^2 - 90.776\left(\frac{\text{Base}}{\text{Acid}}\right)^2 - 273.915(\text{Sr})^2 \\ & R^2 = 0.917,\end{aligned} \tag{18}$$

$$\begin{aligned}\text{FT} = {} & 2431.733 - 2.260\,(\text{SiO}_2) + 13.673\,(\text{Al}_2\text{O}_3) \\ & - 13.201\,(\text{CaO}) - 41.276\,(\text{Na}_2\text{O}) - 15.940\,(\text{K}_2\text{O}) \\ & - 7.826\,(\text{Fe}_2\text{O}_3) - 14.859\,(\text{TiO}_2) - 6.228\,(\text{SO}_3) \\ & - 6.468\left(\frac{\text{Base}}{\text{Acid}}\right) + 0.067(\text{SiO}_2)^2 - 0.020(\text{Al}_2\text{O}_3)^2 \\ & + 0.283(\text{CaO})^2 + 0.437(\text{Na}_2\text{O})^2 - 2.995(\text{K}_2\text{O})^2 \\ & + 0.009(\text{Fe}_2\text{O}_3)^2 - 1.146(\text{TiO}_2)^2 + 0.10(\text{SO}_3)^2 \\ & - 19.668\left(\frac{\text{Base}}{\text{Acid}}\right)^2 \\ & R^2 = 0.941.\end{aligned} \tag{19}$$

The distributions of the differences between values predicted from (17) to (19) and the actual determined amounts of IDT, ST, and FT are shown in Figures 3, 4, and 5, respectively.

4.3. ANFIS Prediction. Input 8, the best predictor in the multivariable regression procedure, was used to determine whether ANFIS is able to predict AFTs more accurately than regression. The determination was conducted by using the MATLAB software package, ANFIS menu, to identify the relationships between the "11 oxide + (Base/Acid) + Sr" input variable and IDT, ST, and FT.

The first step in a neurofuzzy inference system is the determination of system input and output. For this study, input 8 was used as the input to the system. The outputs of the system include three temperatures, IDT, ST, and FT, and each of these outputs has been studied separately.

The fuzzy inference system used in this research was the Sugeno system. The output functions in the Sugeno system are linear or constant. A rule in the fuzzy Sugeno model is:

$$\begin{aligned}&\text{if input 1} = x, \\ &\text{input 2} = y, \text{then the output is } z = ax + by + c.\end{aligned} \tag{20}$$

In the Sugeno system, for a zero-order model, the z plane is constant ($a = b = 0$). The plane of z_i output of any rule is weighted by w_i. The system's final output is the weighted average of all outputs, which is calculated as follows:

$$\text{finaloutput} = \frac{\sum_{i=1}^{N} w_i z_i}{\sum_{i=1}^{N} w_i}. \tag{21}$$

The subtractive clustering scheme was used to cluster data. The subtractive clustering algorithm is a fast, one-pass algorithm for estimating the number of clusters and the cluster centers in a set of data. Subtractive clustering is based on densitometry in space. In this work, the best designed neurofuzzy system for each of the three temperatures was a system with three clusters that could predict the AFTs with the highest correlation coefficients. For IDT, we selected range of influence, squash factor, accept ratio, and reject ratio as 0.5, 1.6, 0.5, and 0.15, respectively. For ST, we selected the values for these same four factors as 0.5, 1.5, 0.5, and 0.15, respectively. For FT, we selected the values of 0.4, 1.5, 0.5, and 0.15, respectively. The structure of the model based on this neurofuzzy system designed for prediction of FT, as an example, is shown in Figure 6. Gaussian membership functions were used in this work. Each of the input variables was assigned to one of three linguistic classes, that is, low, medium, and high. For training of the ANFIS, the hybrid method was used. The data were divided into two subsets. We used 780 sets of data for training, and the remaining 260 sets were used for testing. For the training stage, we selected 200 epochs. After the training was completed, specific rules for predicting each temperature were obtained by the software.

The prediction results achieved by the nurofuzzy system are very promising. The correlation coefficient (R^2), standard deviation (S.D.) of the residual, and residual (% difference between actual and predicted AFTs) less than 120°F (Re < 120°F) for neurofuzzy and nonlinear regression (quadratic) are shown in Table 5. It can be seen that the predictions of AFTs for ANFIS are significantly better than those from multivariable regression.

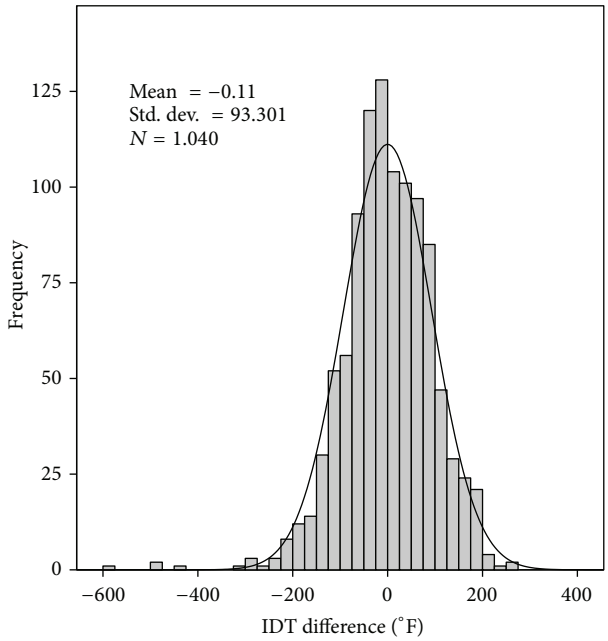

Figure 3: Distribution of difference between actual and estimated IDT values (see, (17)).

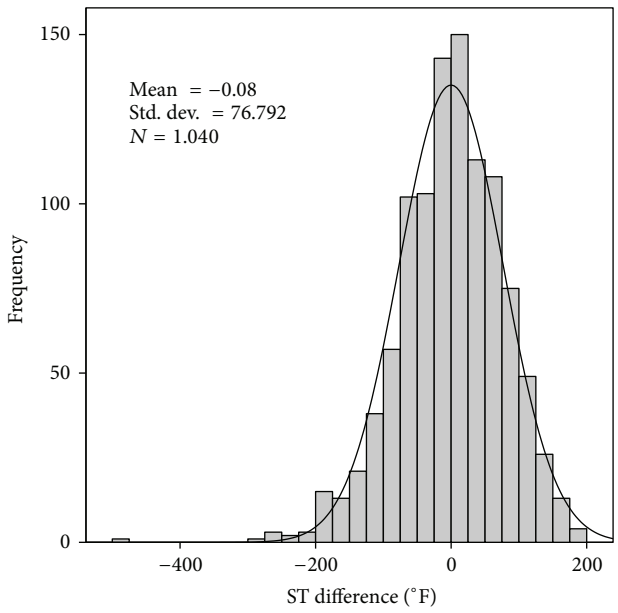

Figure 4: Distribution of difference between actual and estimated ST values (see, (18)).

The IDT estimated by ANFIS in the testing stage compared to actual determined values are shown in Figure 7. The distribution of the differences between actual and estimated IDT is shown in Figure 8.

Figure 9 shows ST estimated by ANFIS compared to actual determined values. The distribution of the differences between actual and estimated ST is shown in Figure 10.

FT estimated by ANFIS in the testing stage compared to actual determined values is shown in Figure 11.

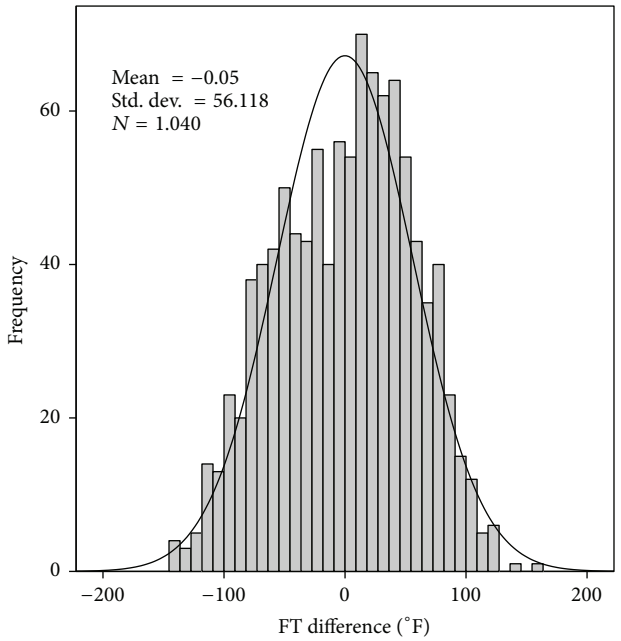

Figure 5: Distribution of difference between actual and estimated FT values (see, (19)).

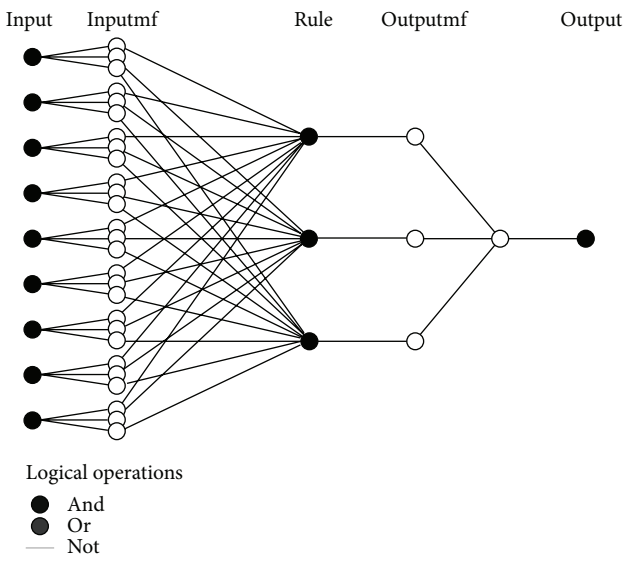

Figure 6: ANFIS model structure for prediction of FT.

The distribution of differences between actual and estimated FT is shown in Figure 12.

4.4. Technical Considerations. Table 4 shows that nonlinear regression analysis produced more accurate results than linear analysis, with regard to correlation coefficient R^2 and percent of residual Re less than 120°F (49°C). An R value of 0.7 is generally acceptable, 0.8 is good, and 0.9 or higher is excellent [8]. In the present study, it can be seen that, for nonlinear correlations (see (17) to (19)), R values reached 0.943 (R^2 of 0.891), 0.957 (R^2 of 0.917), and 0.97 (R^2 of 0.94)

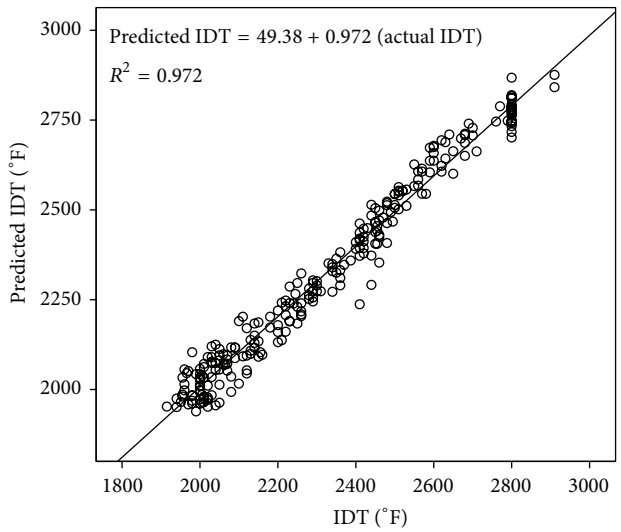

Figure 7: ANFIS estimated IDT in testing stage versus actual determined value.

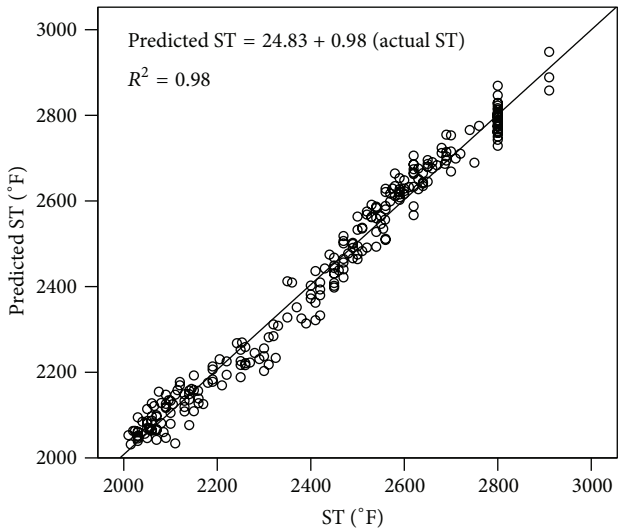

Figure 9: ANFIS estimated ST values in testing stage versus actual determined ST values.

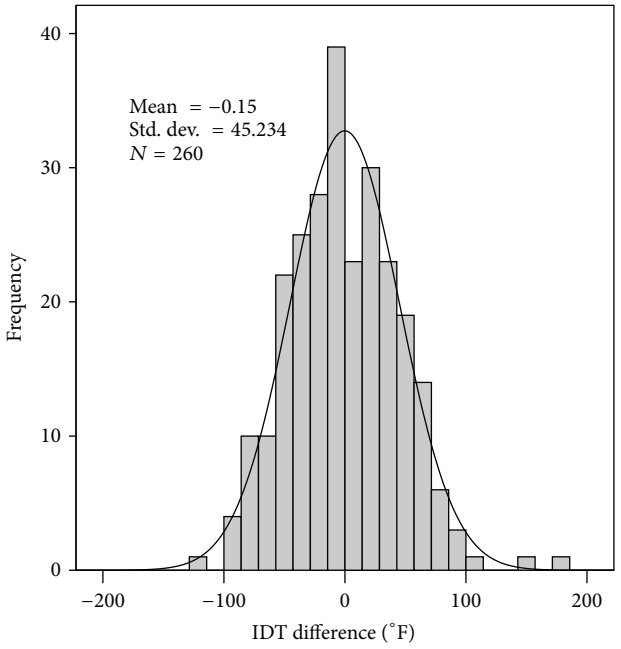

Figure 8: Distribution of difference between actual and estimated IDT values in testing stage.

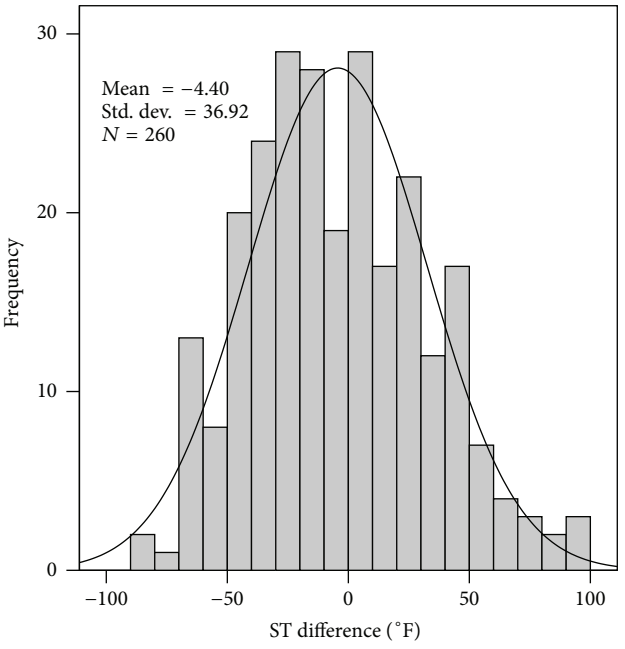

Figure 10: Distribution of difference between actual and estimated ST values in testing stage.

for the prediction of IDT, ST, and FT, respectively; whereas, the ANFIS prediction improved the R values to 0.985, 0.99, and 0.995 for the prediction of IDT, ST, and FT, respectively (Table 5).

As mentioned before, test results from different laboratories for the AFT of a given coal may differ by 20–100°C [4–6]; therefore, the residual can be used as a criterion for the comparison of prediction error and common experimental errors in the laboratory for the regression and ANFIS approaches. In the current work, the nonlinear correlations (see (17) to (19)) can estimate IDT, ST, and FT with a difference less than 120°F (49°C) for about 83, 90, and 98%, respectively. The standard deviations for these nonlinear equations are between 56–93°F (13–34°C) (Input 8, Table 4). The differences less than 120°F for ANFIS prediction of IDT, ST, and FT are 99, 100, and 100%, respectively (Table 5); the standard deviation is between 23 and 45°F (−5 to 7°C) as well (Table 5). These fall within the experimental errors of the measurements. The above-mentioned results indicate that the final proposed nonlinear correlations and ANFIS model were satisfactorily successful; however, the ANFIS prediction is much better than nonlinear correlations.

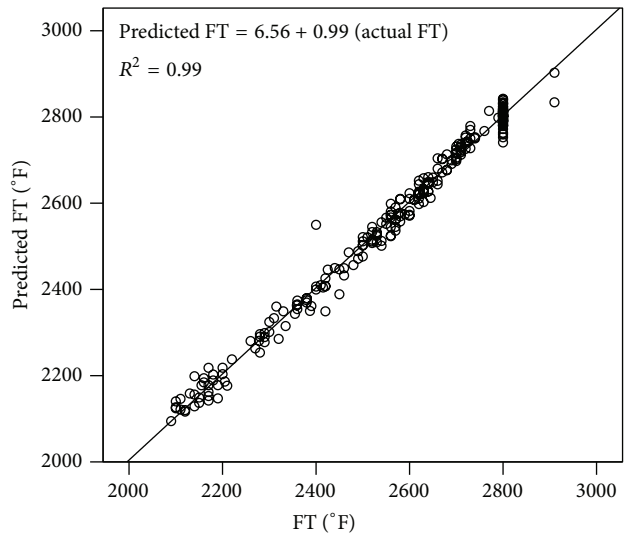

Figure 11: ANFIS estimated FT values in testing stage versus actual determined FT values.

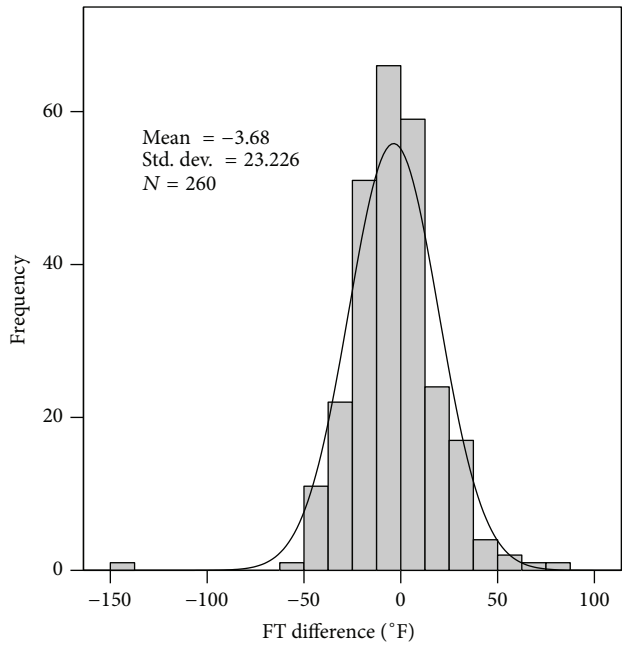

Figure 12: Distribution of difference between actual and estimated FT values in testing stage.

In other works related to this study, G. Özbayoğlu and M. E. Özbayoğlu (2006) applied linear and nonlinear regression analyses for 30 coal samples collected from different parts of Turkey to develop correlations between the chemical composition of ash, ash content, mineral matter content, specific gravity, and Hardgrove index and ash fusion temperatures; the accuracies of R of 0.955, 0.963, and 0.934 were archived for ST, MT (melting temperature), and FT, respectively [11]. Lolja et al. (2002) predicted AFTs for 17 Albanian coals using the chemical compositions of various coal ashes [9]. Seggiani (1999) used a database that included about 300 ash samples from coals of different sources and biomasses. The proposed equations could predict the ash fusion temperatures with correlation R of 0.8 to 0.92 and standard deviation of 45 to 80°C [8].

In the current work, as mentioned before, we used a database that was spread out over a large area, and we used 1040 data sets. Good correlations of R of 0.943, 0.957, and 0.97 in regression and 0.985, 0.99, and 0.995 in ANFIS modeling were achieved for the predictions of IDT, ST, and FT, respectively; this is not in agreement with the Seggiani (1999) suggestion [8] that the statistical approach can give good results in a small number of samples with similar composition. This work shows that a suitable combination of variables can improve the accuracy of the predictions of AFTs, although it is used in widespread databases.

5. Conclusions

(i) Single-variable regression studies show that the SiO_2, Al_2O_3, and Fe_2O_3, as well as base, acid, base/acid, and silica ratios, are significant variables for predicting AFTs in the examined database.

(ii) The basic oxide is the most significant variable with negative effect on AFTs; consequently, the coal ash is favorable with the least basic oxide.

(iii) The acidic oxide is the second significant variable with positive effect on AFTs. Thus, coal ash with a high percentage of acidic oxide is desirable.

(iv) The "11 oxide + (Base/Acid) + Silicate ratio" combination is the best predictor of AFTs in nonlinear equations. This prediction is more precise for FT than for ST or IDT.

(v) The nonlinear equation with "11 oxide + (Base/Acid) + Silicate ratio" input variable can predict IDT, ST, and FT with acceptable correlation coefficients (R^2) of 0.891, 0.917, and 0.94 and standard deviation of about 56–93°F (13–34°C).

(vi) The ANFIS prediction improved the correlation coefficients (R^2) to 0.972, 0.98, and 0.99 for the prediction of IDT, ST, and FT, respectively. Correlation coefficients of these magnitudes have not been reported in previously published works.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgment

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