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Arch. Min. Sci., Vol. 59 (2014), No 4, p. 1061–1076

Electronic version (in color) of this paper is available: http://mining.archives.pl

DOI 10.2478/amsc-2014-0073

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ASSESSMENT OF SELF-HEATING SUSCEPTIBILITY OF INDIAN COAL SEAMS – A NEURAL NETWORK APPROACH

OCENA SKŁONNOŚCI POKŁADÓW WĘGLA W INDIACH DO SAMOZAPŁONU – PODEJŚCIE OPARTE O WYKORZYSTANIE SIECI NEURONOWYCH

The paper addresses an electro-chemical method called wet oxidation potential technique for determining the susceptibility of coal to spontaneous combustion. Altogether 78 coal samples collected from thirteen different mining companies spreading over most of the Indian Coalfields have been used for this experimental investigation and 936 experiments have been carried out by varying different experimental conditions to standardize this method for wider application. Thus for a particular sample 12 experiments of wet oxidation potential method were carried out. The results of wet oxidation potential (WOP) method have been correlated with the intrinsic properties of coal by carrying out proximate, ultimate and petrographic analyses of the coal samples. Correlation studies have been carried out with Design Expert 7.0.0 software. Further, artificial neural network (ANN) analysis was performed to ensure best combination of experimental conditions to be used for obtaining optimum results in this method.

All the above mentioned analysis clearly spelt out that the experimental conditions should be 0.2 N $KMnO_4$ solution with 1 N KOH at 45°C to achieve optimum results for finding out the susceptibility of coal to spontaneous combustion. The results have been validated with Crossing Point Temperature (CPT) data which is widely used in Indian mining scenario.

Keywords: wet oxidation potential, spontaneous heating, correlation studies, artificial neural network analysis, CPT

W pracy omówiono możliwości wykorzystania metody elektro-chemicznej zwanej metodą określania potencjału utleniającego w procesie mokrym do określania skłonności węgla do samozapłonu. Dla potrzeb eksperymentu zebrano 78 próbek węgla z trzynastu kopalni w obrębie Indyjskiego Zagłębia Węglowego. Przeprowadzono 936 eksperymentów, w różnych warunkach prowadzenia procesu aby zapewnić standaryzację metody w celu jej szerszego zastosowania. Dla każdej próbki przeprowadzono 12 eksperymentów metodą badania potencjału utleniającego w procesie mokrym. Wyniki skorelowano z własnościami danego węgla przez przeprowadzenie badania petrograficznych i wytrzymałościowych

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parametrów węgla. Procedurę korelacji wykonano z wykorzystaniem oprogramowania Design Expert 7.0.0, następnie przeprowadzono analizę z wykorzystaniem sieci neuronowych w celu opracowania najkorzystniejszej kombinacji warunków eksperymentu do wykorzystania dla uzyskania optymalnych wyników.

Badania wykazały, że najkorzystniejsze warunki dla procesu to zastosowanie roztworu 0.2 N $KMnO_4$ z 1 N KOH przy 45°C dla uzyskania optymalnych wyników określania skłonności pokładów do samozapłonu. Walidację wyników przeprowadzono w oparciu o wyniki badania metodą określania temperatury przejścia (Crossing Point Temperature), szeroko stosowaną w przemyśle wydobywczym w Indiach.

Słowa kluczowe: metoda badania potencjału utleniającego w procesie mokrym, samozapłon, badanie korelacji, sztuczne sieci neuronowe, temperatura przejścia

1. Introduction

Coal mine fire is inherent to the coal mining industry since its inception. It is caused mainly due to spontaneous combustion of coal (Feng et al., 1973; Kuchta et al., 1980; Yuan & Smith, 2012; Xuyao et al., 2011; Singh et al., 1990). In spontaneous combustion; coal, when exposed to air, undergoes natural oxidation even at ambient temperatures. During the process, it liberates small quantity of heat. If this heat is not dissipated by conduction or convection or radiation or by combination of these three processes, there occurs further rise in temperature, which accelerates the rate of oxygen sorption and production of heat, resulting in fire. Self-heating would be facilitated in conditions where large mass of coal is involved and ventilation is neither too little to restrict coal-oxygen interaction nor too high to dissipate away all the heat generated from above (Banerjee, 2000). It leads to huge loss of coal properties, environmental pollution, hindrance to normal production and productivity, damage to surface structures or adjacent property etc. Thereby, safety and economic aspects of a mine is jeopardized. In one of the study, it has been concluded that China's coal fires, which consume an estimated 20-200 million tons of coal a year, make up as much as 1 percent of the global carbon dioxide emissions from fossil fuels (http://en.wikipedia/wiki/coal_seam_fire). Coal stockpiles are prone to smouldering combustion especially where large quantities are stored for extended periods. Self-heating leading to coal mine fires in underground start in a very small scale and gradually expand in size. It reveals from the careful analysis of occurrence of these fires that most of these fires could have been averted if suitable preventive measures have been taken. The first step for taking preventive measures is to assess the susceptibility of coal seams.

A number of researchers have carried out their studies on self-heating susceptibility of coal seam. It is pertinent to put forward some of their findings here. Nandy et al. (1972) determined crossing point temperature (CPT) of more than 50 Indian coals. Ozdeniz (2010) determined spontaneous combustion in an industrial-scale coal stockpile. Kim and Sohn (2012) proposed new methods to suppress spontaneous ignition of coal stockpiles in a coal storage yard through numerical simulation. Low-temperature oxidation of four different-rank Turkish coals was studied at 40, 60 and 90°C in order to assess the effects of temperature, particle size, coal petrography, and coal rank by monitoring CO_2 and CO formation rates and calculated CO/CO_2 ratios (Baris et al., 2012). Spontaneous combustibility characteristics of Chirimiri Coals were carried out by Pattanaik et al. (2011) and they concluded that degree of proneness to spontaneous combustion increases with the increase of vitrinite and exinite, but decreases with the increase of internite content. Feng et al. (1973) developed liability index of smouldering combustion based on relative ignition temperature, popularly known as FCC index. Ogunsola and Mikula (1990) studied the spontaneous combustion characteristics of four Nigerian coals by determining crossing point temperature and liability index. Mahadevan and Ramlu (1985) devised segmented approach to analyze crossing point temperature curve. They defined an index named as MR index. Peroxy complex formation of coal during oxidation has also been used to evaluate its smouldering combustion susceptibility (Banerjee et al., 1988). Some of the other methods attempted by different researchers are WITS-EHAC liability index (Gouws & Wade, 1989; Gouws & Knoetze, 1995), gas indices study (Singh et al., 2007), development of a device for modeling the behaviour of coal under conditions that may lead to smouldering combustion (Arief & Gillies, 1995), knowledge management system in a longwall mining operations that is liable to smouldering combustion (Singh et al., 1990). Many researchers have tried to correlate the self-heating susceptibility of coal with the intrinsic properties with a great degree of success (Ghosh, 1986; Panigrahi et al., 2000; Kaymakci & Didari, 2002; Beamish & Arisoy, 2008; Sahu et al., 2009). Sahu et al. (2009) applied an empirical approach for classification of coal seams based on their proneness to self-heating. Janusz Cygankiewicz (Cygankiewicz, 2000) determined susceptibility of coals to spontaneous combustion using an adiabatic test method in the Central Mining Institute's Ventilation Division, Katowice, Poland. A prototype test stand has been designed and assembled, the basic element of which was an adiabatic calorimeter. The measurement was carried out from the initial temperature of 40°C up to 185°C. Temperature increase in the adiabatic calorimeter, and the time of reaching 185°C (t) or stoppage considered a basis for classification of coals. On the basis of the results of tests coals were classified into four groups, viz. Low susceptibility: $t > 500$ h; Medium susceptibility: $200 < t \leq 500$ h; High susceptibility: $100 < t \leq 200$ h; Very high susceptibility: $t < 100$ h. Cygankiewicz et al. (2006) made a classification of coals according to their susceptibility to spontaneous combustion on the basis of specific surface area calculated from sorption of carbon dioxide and nitrogen. They carried out this study with 18 bituminous coal samples. They opined that susceptibility to spon-com in the initial stage of the process is limited by the amount of oxygen adsorbed in the macro-pores and meso-pores of a given coal sample. Oxygen accumulated in the macro-pores can diffuse into micro and submicro pores and, in the initial stage of the process, react with organic matter with low energy bonds, located between carbon molecules in carbon chains, and then, with passing time and increasing temperature, interacts with atoms of carbon with higher bond energy. Bituminous coal of higher value of specific surface area has larger potential surface for contact with oxygen, thus interacting with higher number of oxygen molecules and as a result its susceptibility to spontaneous combustion is also higher. Qilin and Shujie (2004) found out rate of oxygen consumption by coal during its self-heating in the laboratory of Shanghai Energy Company Ltd in China. The investigations were carried out for different diameters of grains, at different temperatures and different concentrations of oxygen in the flowing air. The grain distributions were accepted as: 6.5-9 mm, 4.5-6.5 mm, 2.5-4.5 mm, 0.9-2.5 mm and 0.25-0.8 mm. Oxygen content in the inlet air varied from 0.95-21% and temperature in coal reaction chamber changed from 30.1-195.6°C. From the results of the investigations they concluded that oxygen consumption by coal depends on diameter of grain size of coal, the temperature and concentration of oxygen. Skotniczny (2008) discussed the results of three dimensional CFD simulations of temperature and carbon monoxide propagation inside longwall goafs during self-combustion process. Because of high Reynolds number ($Re > 10^5$) he considered turbulent flow in longwall excavations. He expressed the simulation results in a form of three-dimensional distributions of temperature inside goafs and longwall zone, carbon monoxide concentration and as a two-dimensional chart of temperature and carbon monoxide in selected places of goaf-longwall complex.

In addition, very few researchers carried out wet oxidation potential experiments to predict spontaneous combustion characteristics of coal. Tarafdar and Guha (1989) conducted wet oxidation experiments with seven coal samples. They observed that higher the potential difference more susceptible would be the coal towards smouldering combustion. Panigrahi et al. (2004) conducted experiments with 12 coal samples from Indian Coalfields. They suggested that the wet oxidation potential method has the potentiality to predict susceptibility to self-heating of coal more accurately than CPT.

The present paper aims to assess the self-heating susceptibility of coal through extensive application of wet oxidation potential method with different experimental conditions and standardize this method by neural network analysis utilizing data obtained from proximate, ultimate and petrographic analyses.

2. Experimental investigation

Seventy eight (78) coal samples covering fiery and non-fiery seams of thirteen mining companies of India were collected for this investigation. The companies are Eastern Coalfields Ltd. (ECL), Bharat Coking Coal Ltd. (BCCL), Central Coalfields Ltd. (CCL), Mahanadi Coalfields Ltd. (MCL), South Eastern Coalfields Ltd. (SECL), Northern Coalfields Ltd. (NCL), Western Coalfields Ltd. (WCL), North Eastern Coalfields (NEC), Singareni Collieries Company Ltd. (SCCL), IISCO Steel Plant (ISP) SAIL, Monnet Ispat & Energy Ltd., Neyveli Lignite Corporation (NLC) and Tata Steel Ltd. Location of collected coal samples extends over Raniganj, Jharia, Karanpura, Ramgarh, Bokaro, Ib valley, Raigarh, Chirimiri, Son valley, Wardha valley, Kamptee, Singrauli, Pranhita-Godavari valley, Bikaner, Neyveli and Makum coal basins. Details of coal samples, i.e. sample code, name of the mine and the seam and mining company is presented in Table 1.

The coal samples were collected following channel sampling procedure (Peters, 1978; Indian Standard: 436, 1964) and brought to the laboratory in sealed condition for analysis. Samples were ground and sieved to suitable size (-212 μ) making necessary coning and quartering. Samples were prepared for various analyses namely proximate (moisture, volatile matter, ash and fixed carbon), ultimate (carbon, hydrogen, nitrogen, sulphur and oxygen) and petrographic analyses (vitrinite, inertinite, liptinite and mineral matter content). The proximate, ultimate and petrographic analyses were carried out following standard procedures (Indian Standard: 1350, Part-I, 1969; ASTM D 5373-93, 1993; ICCP: 1971, ICCP: 1994, Indian Standard: 9127, Part -I, 1979, Indian Standard: 9127, Part -II, 1979).

2.1. Wet oxidation potential analysis

Wet oxidation potential analysis was carried out with potassium permanganate ($KMnO_4$) as oxidizer in potassium hydroxide (KOH) solution. Equivalence factor of $KMnO_4$ in this case was maintained as 158.04/3 = 52.68. Experiments were carried out at different concentration of $KMnO_4$, viz. 0.05N, 0.1N, 0.15N and 0.2N in 1 N KOH and at 27, 40 and 45°C. Thus, 12 experiments were carried out for a particular coal sample. In total, 936 experiments were carried out for wet oxidation potential analysis. 100 ml of such mixture was taken in a beaker and a calomel reference electrode and a carbon electrode was immersed in it. The potential difference, i.e. EMF, in mV, was measured between these electrodes by using a multimeter after attaining a stable reading. 0.5 g of coal sample of -212 μ size was added in this mixture and was continuously

TABLE 1

Details of coal samples chosen for experiments

S No.	Mine/company	Seam	S No.	Mine/company	Seam
1	Central Kajora, ECL	RVIII	40	Churcha East, SECL	V
2	Parascole East, ECL	RVII	41	Haldibari, SECL	XB
3	Shamsunderpur, ECL	RVII	42	Kamptee OCP, WCL	VB
4	Lakhimata, ECL	Metadih	43	Saoner Mine 1, WCL	IV (M)
5	Lakhimata, ECL	BII	44	Saoner Mine 3, WCL	V
6	Jhanjra, ECL	RVIIA	45	Umrer OCP, WCL	IV
7	MIC unit Jhanjra, ECL	RVI	46	New Majri III, WCL	Majri
8	Jhanjra, ECL	RVII	47	Ghuggus OCP, WCL	Meyo Bottom
9	Kottadih Project, ECL	RV	48	Ghuggus OCP, WCL	Meyo Middle
10	Kottadih, ECL	RIII/II	49	Naigaon OCP, WCL	Meyo Bottom
11	Khaskajora, ECL	RVIIIA	50	Naigaon OCP, WCL	Meyo Middle
12	Khaskajora, ECL	RVIIIB	51	Jhingurda, WCL	Jhingurdah
13	Kumardhubi, ECL	Singpur Top	52	Jayant OCP, WCL	Turra
14	Bansdeopur, BCCL	VIII	53	Jayant OCP, WCL	Purewa Bottom
15	Victoria West, BCCL	Ramnagar	54	Jayant OCP, WCL	Purewa Top
16	Sudamdih shaft, BCCL	XI/XII	55	Amlohri OCP, WCL	Purewa Merge
17	Bastacolla, BCCL	0	56	Amlohri OCP, WCL	Turra
18	Bastacolla, BCCL	I	57	Tipong, NEC	20 Feet
19	Bastacolla, BCCL	II	58	Tipong, NEC	60 Feet (Bottom)
20	Moonidih, BCCL	XVI (T)	59	Tipong, NEC	60 Feet (Top)
21	Mudidih, BCCL	IX	60	Tirap OCP, NEC	8 Feet
22	Kalyani, CCL	Karo (Major)	61	Tirap OCP, NEC	60 Feet (Top)
23	Argada, CCL	I	62	Tirap OCP, NEC	20 Feet
24	Argada, CCL	J	63	Kakatiya LW, SCCL	I
25	Hesagora, CCL	X Bottom	64	Kakatiya LW, SCCL	IA
26	Churi, CCL	Lower Bachra	65	Kakatiya LW, SCCL	II
27	KD Hessalong, CCL	Dakra	66	Kakatiya LW, SCCL	III
28	Kuju, CCL	VII	67	Adriyala Shaft, SCCL	I
29	Lilari OCP, MCL	Lajkura Top	68	RK New Tech incline, SCCL	1A
30	Belpahar OCM, MCL	IB	69	Sijua, Tata Steel Ltd.	XIII
31	Belpahar OCM, MCL	Rampur Top	70	Sijua, Tata Steel Ltd.	XIV
32	Belpahar OCM, MCL	Rampur Bottom	71	6&7 Pits, Tata Steel Ltd.	IX
33	Lakhanpur OCP, MCL	Lajkura Top	72	6&7 Pits, Tata Steel Ltd.	XI
34	Jagannath OCP, MCL	III	73	Milupara, Monnet Ispat & Energy Ltd.	II
35	Anjan Hill, SECL	III	74	Kondkel, Monnet Ispat & Energy Ltd.	III
36	NCPH, SECL	III	75	Chasnalla, ISP, SAIL	XII
37	Rajnagar RO, SECL	$8A_2$	76	Western Quarry, ISP, SAIL	XIII/XIV
38	5&6 Incline, SECL	Index	77	Neyveli Mine 1A, NLC	Lignite
39	Churcha West, SECL	V	78	Barsingsar lignite, NLC	Lignite

stirred using a magnetic stirrer. The potential difference was recorded over a period of time till a nearly constant value was attained. Temperature of the mixture was measured with a calibrated temperature recorder. Potential difference and temperature were recorded at an interval of 1 min. Each experiment takes about an hour. The photograph for wet oxidation potential method is shown in Fig. 1. The difference between potential difference (PD) of the mixture before adding coal sample and after complete oxidization of coal sample was calculated for each sample and total time taken for each experiment was recorded and thus rate of reduction of potential difference was calculated. This rate (RPD) was considered as a parameter for susceptibility of coal towards smouldering combustion and expressed in mV/min.

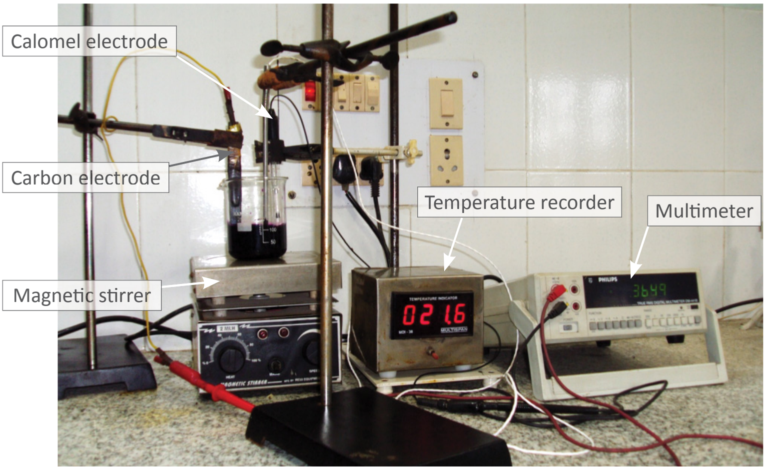


Fig. 1. Photograph showing experimental set up for wet oxidation potential method

3. Results

Table 2 represents independent and dependent variables. Independent variables are moisture, volatile matter (VM), carbon (C), hydrogen (H), nitrogen (N), sulphur (S) and oxygen (O) on dry ash free basis; wt. %, and maceral content, viz. vitrinite (V), inertinite (I) and liptinite (L) on mineral matter free basis, vol. %. Dependent variables are rate of reduction of potential difference (RPD) in different experimental conditions. Acronyms used for rate of reduction of potential difference (RPD) in different experimental conditions are as follows:

RPD1 – RPD at 0.05N $KMnO_4$ with 1 N KOH solution at 27°C temperature,
RPD2 – RPD at 0.05N $KMnO_4$ with 1 N KOH solution at 40°C temperature,
RPD3 – RPD at 0.05N $KMnO_4$ with 1 N KOH solution at 45°C temperature,
RPD4 – RPD at 0.1N $KMnO_4$ with 1 N KOH solution at 27°C temperature,
RPD5 – RPD at 0.1N $KMnO_4$ with 1 N KOH solution at 40°C temperature,
RPD6 – RPD at 0.1N $KMnO_4$ with 1 N KOH solution at 45°C temperature,
RPD7 – RPD at 0.15N $KMnO_4$ with 1 N KOH solution at 27°C temperature,
RPD8 – RPD at 0.15N $KMnO_4$ with 1 N KOH solution at 40°C temperature,

RPD9 – RPD at 0.15N $KMnO_4$ with 1 N KOH solution at 45°C temperature,
RPD10 – RPD at 0.2N $KMnO_4$ with 1 N KOH solution at 27°C temperature,
RPD11 – RPD at 0.2N $KMnO_4$ with 1 N KOH solution at 40°C temperature,
RPD12 – RPD at 0.2N $KMnO_4$ with 1 N KOH solution at 45°C temperature.

Results of independent and dependent variables of 78 coal samples are shown in statistical form in Table 2. Minimum, maximum, mean and standard deviations of all variables are depicted. In order to determine the best combinations of experimental conditions to achieve optimum results in wet oxidation potential method, results are first analysed statistically and then by Artificial Neural Network analysis.

Repeatability of experimental results was verified with three coal samples, carrying out 5 experiments each for a sample. Sample No. 13 (Kumardhubi Colliery, Singhpur top seam, ECL) with 0.2N $KMnO_4$ in 1N KOH solution at 40°C gives PD of 62, 61.1, 60.5, 61.7, 60.9 mV whereas Sample No. 16 (Sudamdih Shaft Mine, XI/XII seam, BCCL) with 0.1N $KMnO_4$ in 1N KOH solution at 27°C shows PD of 48.1, 47.6, 47.1, 47.4, 47.0 mV. Further, with 0.2N $KMnO_4$ in 1N KOH solution at 40°C, Sample No. 46 (New Majri III, Majri Seam) gives PD of 121.3, 119.7, 120.5, 118.5, 119.1 mV. So, standard deviation of these three samples comes out to be 0.54, 0.39 and 0.99 respectively. The coefficients of variation of aforementioned samples are calculated as 0.0088, 0.0082 and 0.0083 respectively.

TABLE 2

Variables and their units obtained from different experimental investigation

Variable	Unit	Minimum	Maximum	Mean	Std. dev.
1	2	3	4	5	6
Independent variables					
Moisture	%	0.4	15.71	5.20	4.08
Ash	%	0.71	38.46	16.18	8.84
Volatile Matter	%	18.62	55.22	38.44	8.13
Fixed Carbon	%	27.94	70.28	48.52	9.17
Carbon	%	70.02	89.56	80.62	4.58
Hydrogen	%	4.73	8.25	6.10	0.84
Nitrogen	%	0.95	2.54	1.76	0.33
Sulphur	%	0.3	7.18	1.02	1.06
Oxygen	%	1.16	19.12	10.44	4.01
Vitrinite	%	21.51	84.79	64.32	11.69
Inertinite	%	8.69	78.49	31.33	12.34
Liptinite	%	0	33.76	4.24	4.60
Dependent variables					
Crossing Point Temperature	°C	125	179	141.74	12.03
RPD1	mV/min.	0.84	9.08	3.36	2.08
RPD2	mV/min.	1.19	12.56	4.61	2.73
RPD3	mV/min.	1.14	13.13	4.77	2.99
RPD4	mV/min.	0.76	10.73	3.74	2.43
RPD5	mV/min.	0.79	14.77	5.14	3.10
RPD6	mV/min.	0.92	14.49	5.53	3.29

1	2	3	4	5	6
RPD7	mV/min.	0.83	10.82	4.04	2.47
RPD8	mV/min.	1.11	16.31	5.62	3.37
RPD9	mV/min.	0.92	14.65	6.01	3.65
RPD10	mV/min.	0.89	12.06	4.35	2.70
RPD11	mV/min.	0.93	15.66	6.24	3.85
RPD12	mV/min.	1.3	19.03	6.82	4.18

Volatile Matter, Carbon, Hydrogen, Oxygen are on dry ash free bases and Vitrinite, Inertinite and Liptinite are on mineral matter free bases

4. Discussion

The results are first analysed statistically by Design Expert 7.0.0 software and then by artificial neural network analysis.

4.1. Correlation between susceptibility indices and intrinsic properties

The correlation studies have been carried out between the different susceptibility indices and the coal characteristics as obtained from proximate, ultimate and petrographic analyses. Design Expert 7.0.0 software was used for statistical analysis. The susceptibility indices are taken as dependent variables and each constituent obtained from the proximate, ultimate and petrographic analyses as independent variables. Volatile matter, carbon, hydrogen and oxygen are taken on dry ash free (daf) bases. Vitrinite and inertinite are considered on mineral matter free bases. The correlation coefficients obtained in all cases are presented in Table 3. It is revealed from Table 3 that RPD12 gives maximum correlation coefficient with moisture, carbon, oxygen and inertinite content. Statistical analyses indicate that with the increase of moisture, volatile matter, oxygen and hydrogen susceptibility to spontaneous heating of coal increases. It has been observed that susceptibility of coal decreases with increase in carbon per cent (on daf basis). Due to presence of large amount of micropores (nanopores) in vitrinite it absorbs oxygen readily and abundantly using less activation energy per unit area and thus experience accelerated oxidation in the earlier stage of self heating (Misra and Singh, 1994). Moreover, non-aromatic molecular structures are common in vitrinite macerals which is responsible for low temperature oxidation. As vitrinite increases, RPD also increases. Correlation coefficients between these two parameters are found to vary from 0.30 to 0.39. Intertinite macerals have dense aromatic molecular structures and higher activation energy involved in it and thus these are not easy prey to oxidation in initial stage. As inertinite increases, RPD decreases. They maintain inverse proportionality and correlation coefficients between these two parameters are found to be fair (r = –0.32 to –0.46). Subsequently, considering M and VM_{daf} as two independent variables, relationships are established with RPD. Best correlation is observed with RPD10, RPD11 and RPD12 (r = 0.94) (Table 3). When O_{daf} and H_{daf} are considered as two independent variables and relationship is established with RPD, best correlation is observed with RPD12 (r = 0.80) (Table 3). Statistical analysis reveals that RPD with ash, vitrinite or inertinite content shows poor correlation so these parameters are not taken into consideration in the artificial neural network analysis.

TABLE 3

Correlation coefficients between different susceptibility indices and results of proximate, ultimate and petrographic analyses

Sl. No.	Intrinsic Characte-ristics	Susceptibility indices											
		RPD1	**RPD2**	**RPD3**	**RPD4**	**RPD5**	**RPD6**	**RPD7**	**RPD8**	**RPD9**	**RPD10**	**RPD11**	**RPD12**
1	M	0.82	0.83	0.81	0.86	0.83	0.81	0.93	0.90	0.90	0.94	0.93	0.94
2	VM	0.80	0.78	0.76	0.76	0.70	0.69	0.77	0.68	0.67	0.75	0.74	0.74
3	A	0.26	0.22	0.20	0.31	0.32	0.35	0.34	0.30	0.32	0.32	0.30	0.29
4	C	0.79	0.76	0.73	0.77	0.70	0.69	0.77	0.69	0.67	0.78	0.76	0.79
5	H	0.71	0.69	0.67	0.65	0.65	0.57	0.62	0.60	0.57	0.61	0.64	0.65
6	O	0.76	0.75	0.72	0.73	0.68	0.68	0.76	0.67	0.65	0.76	0.76	0.77
7	V	0.31	0.35	0.32	0.39	0.45	0.32	0.37	0.30	0.39	0.32	0.34	0.34
8	I	0.33	0.39	0.32	0.41	0.46	0.40	0.46	0.38	0.40	0.39	0.41	0.46
9	M & VM	0.87	0.88	0.85	0.89	0.87	0.85	0.94	0.91	0.90	0.94	0.94	0.94
10	O & H	0.78	0.78	0.77	0.71	0.69	0.67	0.71	0.69	0.64	0.76	0.75	0.80

VM, C, H, O are on daf bases and V and I are on mmf bases

4.2. Application of artificial neural network

In the present study Levenberg – Marquardt (LM) Backpropagation (BP) algorithm (trainlm) is used. This is the most widely used optimization algorithm. MATLAB R2009b version 7.9.0 is used for this neural network analysis. Among simulation tool functions in the neural network of MATLAB, LM is taken as default training function in the BP neural network.

The LM algorithm is an iterative technique that locates a local minimum of a multivariate function that is expressed as the sum of squares of several non-linear, real-valued functions. It has become a standard technique for non linear least square problems, widely adopted in various disciplines for dealing data fitting applications. Levenberg-Marquardt curve-fitting method is actually a combination of two minimization methods the gradient descent method and the Gauss-Newton method. The following steps are followed in the training process using LM algorithm (Fig. 2): where, w_k is the current weight, w_{k+1} is the next weight, E_{k+1} is the current total error, and E_k is the last total error (Hao and Wilamowski, 2010).

i. With the initial weights (randomly generated) the total error (SSE) is evaluated.
ii. An update is made as directed by equation $w_{k+1} = w_k - (J_k^T J_k + \mu I)^{-1} J_k e_k$ where, J is Jacobian matrix, I is the identity matrix, μ is always positive and called combination coefficient.
iii. With the new weights the total error is evaluated.
iv. There could be two possibilities as a result of update. If the current total error is increased then the step is repeated after resetting the weight vector to the previous value and increase combination coefficient μ by a factor of 10 or by some other factors. Then go to step ii and try an update again.
v. If the current total error is decreased then accept the step such as keep the new weight vector as the current one and decrease the combination coefficient μ by a factor of 10 or by same factor as step iv.
vi. Go to step ii with the new weights until the current total error is smaller than the required value.

In the present investigations, physical and chemical characteristics of coal, viz. moisture, volatile matter (daf basis), carbon, hydrogen, oxygen on daf bases were considered as input pa-

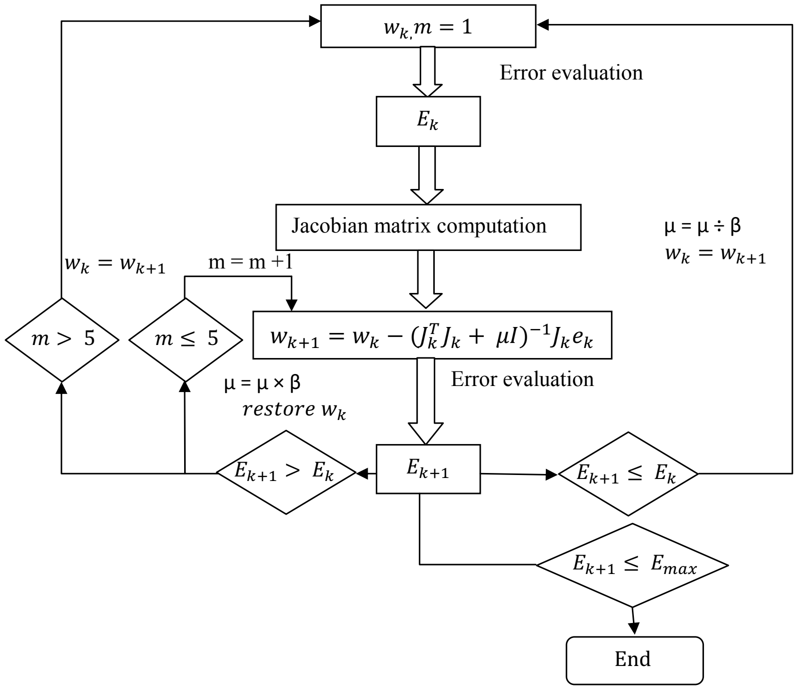


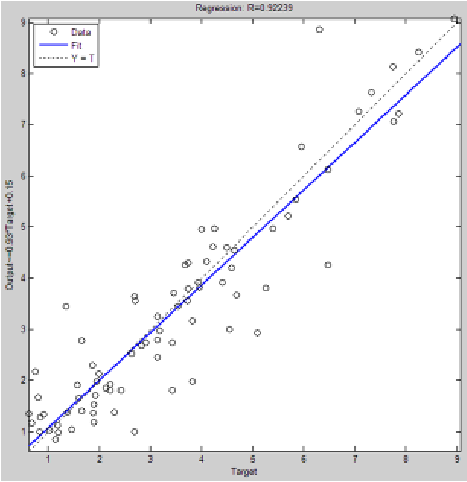
Fig. 2. Block diagram for training using Levenberg-Marquardt algorithm

rameters as these parameters show better correlation coefficient in statistical analysis. The rate of potential difference at different experimental conditions, viz. RPD1, RPD2, RPD3, RPD4, RPD5, RPD6, RPD7, RPD8, RPD9, RPD10, RPD11, and RPD12 were considered as output parameter one at a time. The neural network was run, i.e. training of data with Levenberg – Marquardt (LM) Backpropagation (BP) algorithm till the time maximum correlation coefficient was achieved. Out of 78 data sets, 62 were considered for training, 8 (10% of total data) were considered for validation and remaining 8 data sets (10% of total data) were used for testing. For each output parameter, this exercise was conducted and maximum correlation coefficient (R) and mean squared error (MSE) were recorded. Table 4 depicts the results of analysis from artificial neural network and Fig. 3 gives regression plot for different susceptibility indices. It is revealed from Table 4 and Fig. 3 that RPD12 gives maximum correlation coefficient of 0.96 and minimum mean squared error of 1.51. Therefore by using aforementioned intrinsic properties of coal, viz. moisture, volatile matter, carbon, hydrogen and oxygen, the susceptibility index, RPD12, can also be accurately predicted by artificial neural network.

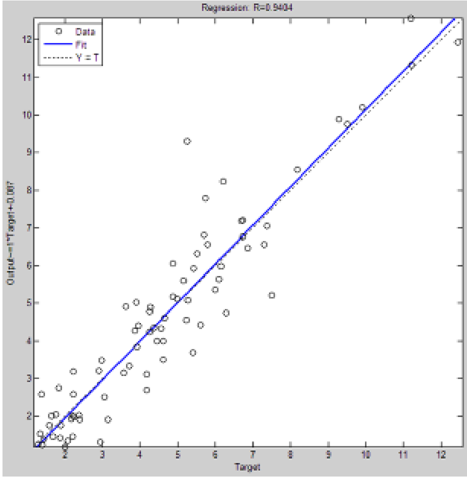
TABLE 4

Results of analysis from artificial neural network

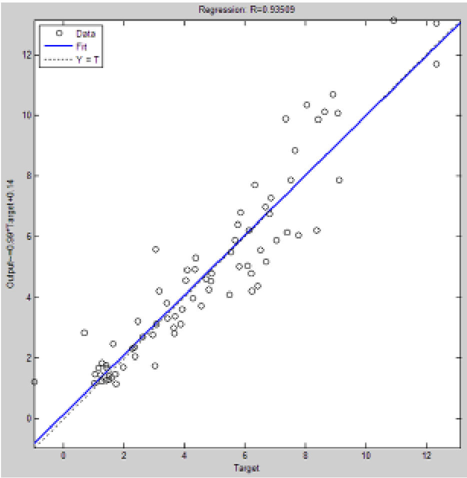
Indices	**RPD1**	**RPD2**	**RPD3**	**RPD4**	**RPD5**	**RPD6**	**RPD7**	**RPD8**	**RPD9**	**RPD10**	**RPD11**	**RPD12**
R	0.92	0.94	0.93	0.91	0.91	0.87	0.9	0.91	0.92	0.93	0.93	0.96
MSE	0.66	0.85	1.11	0.94	1.66	2.65	1.2	2.07	2	1	1.8	1.51



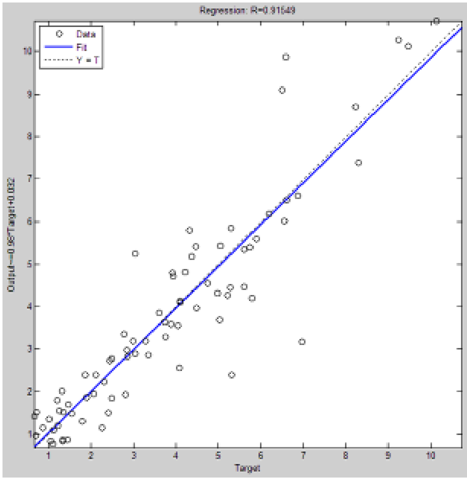
(a) Regression plot for RPD1



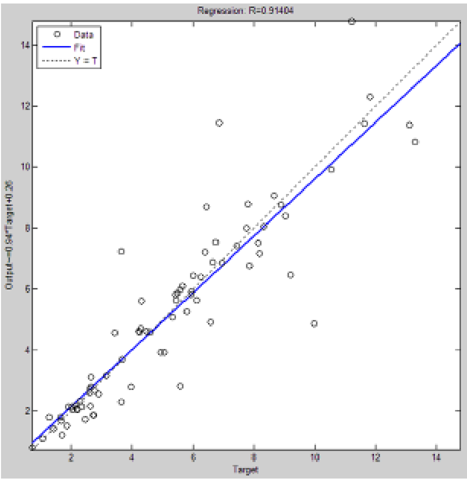
(b) Regression plot for RPD2



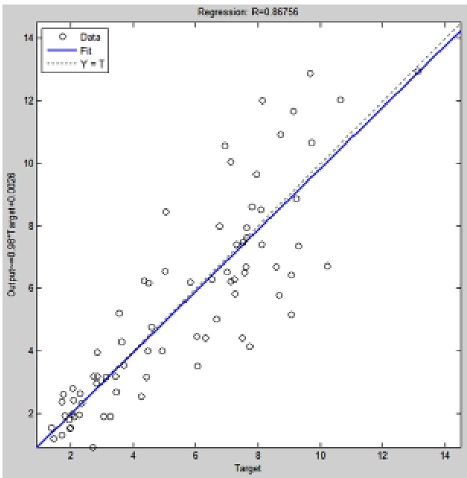
(c) Regression plot for RPD3



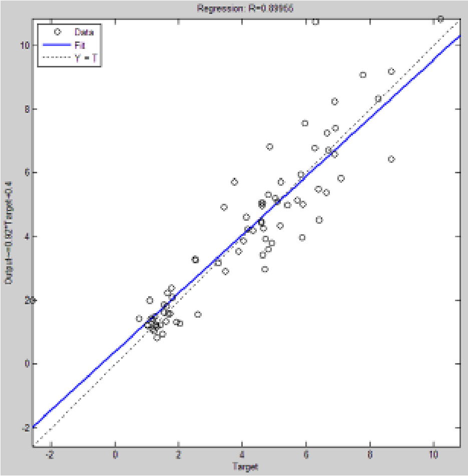
(d) Regression plot for RPD4



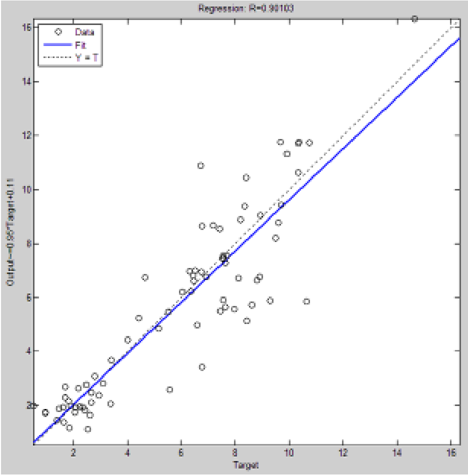
(e) Regression plot for RPD5



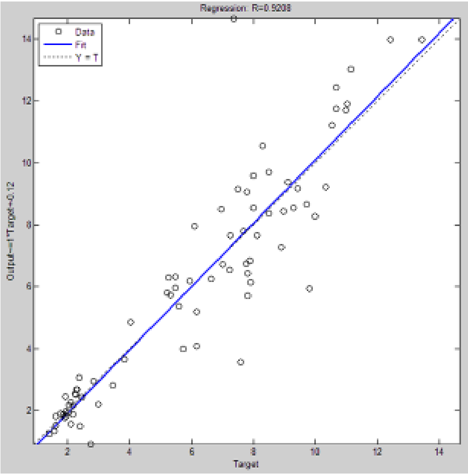
(f) Regression plot for RPD6



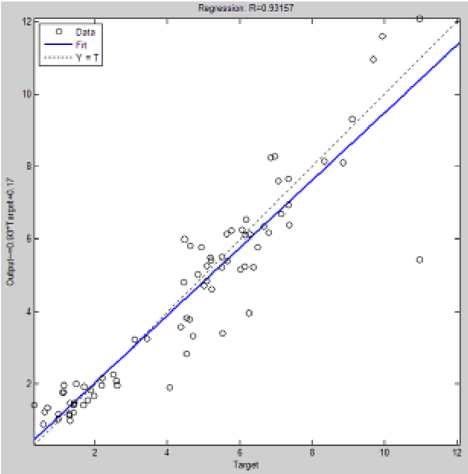
(g) Regression plot for RPD7



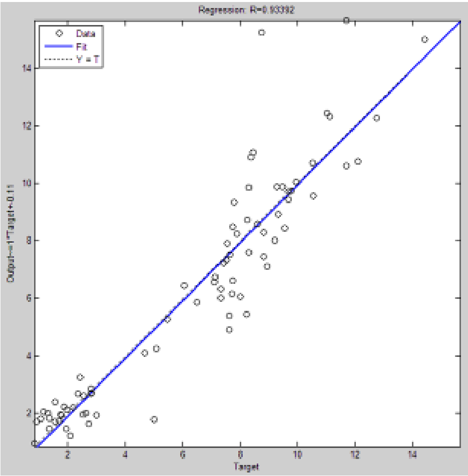
(h) Regression plot for RPD8



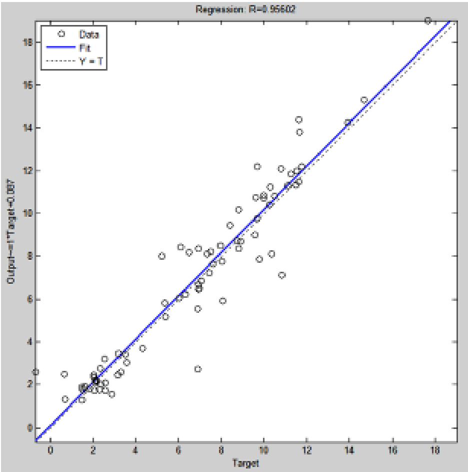
(i) Regression plot for RPD9



(j) Regression plot for RPD10



(k) Regression plot for RPD11



(l) Regression plot for RPD12

Fig. 3. Regression plot for susceptibility indices

5. Validation of results of wet oxidation with CPT

Crossing point temperature (CPT) of 78 coal samples were determined by standard procedure as practiced in India (Panigrahi et al., 1999). Statistical analysis of CPT with RPD12 are carried out with Design Expert 7.0.0 software and represented in Fig. 4 and correlation coefficient value and SD (Standard Deviation) in this case are found to be 0.50 and 3.90 respectively.

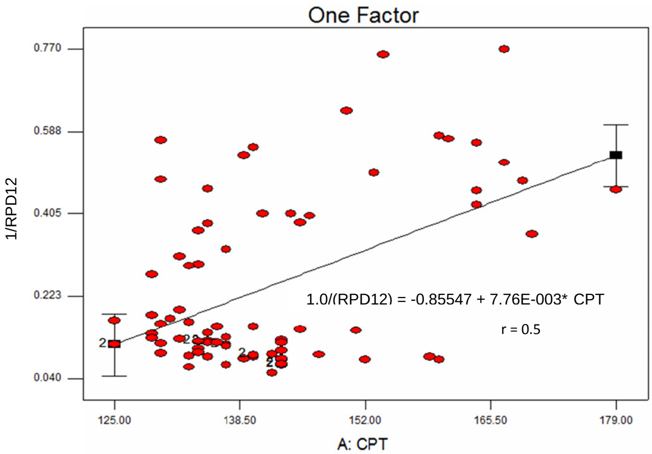


Fig. 4. Correlation of RPD12 with Crossing Point Temperature

5.1. Comparison between RPD12 and CPT

Correlation study with respect to CPT analyses is carried out for comparison purpose. It reveals from Table 3 that poor correlation coefficients are observed when susceptibility index (RPD) is correlated with petrographic constituents. Therefore, these are not considered for comparison purpose. Table 5 details out the comparison between CPT and RPD12 analyses.

TABLE 5

Correlation coefficients obtained from CPT and RPD12 analysis

Sl. No.	Intrinsic Characteristics	Susceptibility indices	
		CPT	RPD12
1	M	0.62	0.94
2	VM_{daf}	0.78	0.74
3	C_{daf}	0.77	0.79
4	H_{daf}	0.62	0.65
5	O_{daf}	0.74	0.77
6	M & VM_{daf}	0.85	0.94
7	O_{daf} & H_{daf}	0.75	0.80

The following points are noteworthy while critically analyzing Table 5:

- Overall improvement of correlation coefficients is noticed while analysis is done with RPD12.
- Maximum improvement in correlation coefficient is observed while making correlation with moisture. In case of RPD12 it is 0.94 while in case of CPT it is 0.62.
- While carrying out correlation studies with C_{daf}, correlation coefficient r is found to be 0.77 in case of CPT analysis and RPD12 analysis depicts its value 0.79.
- Improvement in correlation coefficient is also observed in case of H_{daf} and O_{daf}. While correlation is done with hydrogen, CPT analysis shows r is 0.62 and RPD12 analysis shows it is 0.65. Similarly, when correlation is done with oxygen, CPT analysis shows r is 0.74 and RPD12 analysis shows it is 0.77.
- Correlation coefficient r is also obtained combining M and VM_{daf} for both the analyses. CPT analysis depicts its value 0.85 and RPD12 analysis gives its value 0.94.
- Considering O_{daf} and H_{daf} as two independent variables correlation analysis is made with CPT as well as RPD12. In case of CPT, r is found to be 0.75 and in case of RPD12, its value is 0.80.
- Both the analysis results indicate that RPD12 gives highest correlation coefficient in maximum number of cases.

Keeping the above detailed comparative analysis in view, it is recommended that rate of reduction of potential difference (RPD12) in WOP method using 0.2N $KMnO_4$ solution with 1N KOH at 45°C should be used for determining the susceptibility of coal to spontaneous combustion.

6. Conclusions

The following conclusions may be drawn from the present investigation:

1. Wet oxidation potential technique, an electro-chemical method, is found to be accurate, simple and faster method to determine self-heating characteristics of coal.
2. Amongst the twelve different combinations of WOP method, RPD12, i.e. at 0.2N $KMnO_4$ solution with 1N KOH at 45°C gives higher order correlation coefficient with intrinsic properties of coal, viz. M, C_{daf}, O_{daf} and I_{mmf}. Therefore, 0.2N $KMnO_4$ with 1N KOH solution at 45°C be appropriate one for carrying out wet oxidation potential technique to characterize the spontaneous heating of coal.
3. Applying artificial neural network with the different susceptibility indices and the coal characteristics as obtained from proximate and ultimate analyses, it has been found that RPD12 gives maximum correlation coefficient of 0.96 and minimum mean squared error of 1.51. Therefore it is recommended that WOP method using 0.2N $KMnO_4$ solution with 1N KOH at 45°C should be used for determining the susceptibility of coal to spontaneous combustion.
4. Statistical analysis of CPT with RPD12 shows that RPD12 bear a good correlation with CPT having r value 0.50.
5. A comparison between correlation coefficients obtained from CPT and RPD12 analysis indicates that RPD12 gives highest correlation in maximum number of cases. So, RPD12 is a better susceptibility index than CPT.

Keeping all the observations and analysis in mind, it may be concluded that wet oxidation potential analysis is a suitable method for determining self-heating susceptibility of coal and experimental conditions should be 0.2N $KMnO_4$ solution with 1N KOH at 45°C. However, experimental investigations of different coals of other countries may be carried out to further validate and strengthen WOP method for wider application in coal mining industry of the world.

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Received: 30 September 2013