Trends in Applied Sciences Research

ISSN 1819-3579

Academic
Journals Inc.

www.academicjournals.com

Trends in Applied Sciences Research 7 (6): 434-444, 2012
ISSN 1819-3579 / DOI: 10.3923/tasr.2012.434.444

Structural Characterization of Selected Indian Coals by X-ray Diffraction and Spectroscopic Techniques

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ABSTRACT

The structural parameters of three Indian coals were determined by X-ray diffraction, Raman, UV-Vis-NIR, FTIR spectroscopy and SEM-EDS. This study reveals that the coals contain crystalline carbon of turbo- stratic structure with some amount of amorphous carbon. The average lateral sizes (L_a), stacking heights (L_c) and interlayer spacing (d_{002}) of the crystallite structures calculated from the X-ray intensities range from 28.15-38.43 A°, 22.64-20.16A° and 3.52-3.34 A°, respectively which is higher than that of pure graphite, suggesting a low degree of crystalline order in the studied sample. The decrease in La with rank shows that the carbon in coal is nano crystalline in structure. Raman spectroscopic study confirms the presence of multi layer formation of graphite layer. The defect band at 1355 cm^{-1} is due to benzene or condensed benzene rings present in amorphous carbon. The FTIR spectra of the coals show the presence of stretching vibrations of -OH bonds, aliphatic -CH, -CH_2 and -CH_3 absorptions, C = C and -CH of aromatic structure. The C-H bending frequencies have higher intensity than the C-H stretching band. This is due to the intense broad absorption produced in this region by graphite components. A strong linear relationship exists between the coal structural parameters (fa,d_{002},Lc) and the elemental carbon and volatile contents of the coal which reflects the dependency of the coals structure on their ranks. The SEM image shows the presence of layered structure on the surface. The EDS analysis confirms the presence of Carbon and Oxygen on the surface. The absorption maximum of benzene-oxygen system was found between 235-270 nm. The weak band at the 680 nm is attributed to the $\prod$-$\prod^*$ electronic transitions of the polynuclear aromatic hydrocarbons.

Key words: Turbostratic structure, graphite layers, raman spectroscopy, characterization, coal

INTRODUCTION

The Structural characterization of coal has received much attention among researchers because of its effects on chemical reactivity in combustion, pyrolysis, liquefaction and gasification processes. An awareness of coal structural parameters is essential in predicting and controlling theses processes. Coal structure being complex and heterogeneous makes it structural investigation a topic of serious research among chemist. Coal contains primary macromolecules of polyaromatic-polynuclear structure with some heteroatom group. In the secondary network of coal, aromatic ring-stacking, aliphatic side-chain entanglement, hydrogen bonds, cation bridges and charge-transfer interactions through oxygen functional groups are present along with mineral functional groups. Various instrumental techniques, like TEM (Rouzaud and Obelin, 1990), NMR (Behera *et al*., 2008; Solum *et al*., 2001), FTIR (Manoj and Kunjomana, 2010a), SEM-EDS

(Wu and Steel, 2007), Raman Spectroscopy (Friedel and Carlson, 1972; Manoj and Kunjomana, 2010b; Manoj and Jose, 2011; Li *et al.*, 2006), XRD (Takgi *et al.*, 2004; Sonibare *et al.*, 2010), UV-Vis-NIR (Friedel and Carlson, 1972) have been applied to the investigation of the chemical structure of coal.

X-Ray Diffraction (XRD) is a fundamental characterization technique, that has been established for the determination of structural parameters in carbonaceous materials in coal (Takgi *et al.*, 2004; Sonibare *et al.*, 2010). The aromaticity (fa), interlayer spacing of the crystalline structure (d_{002}) and crystallite sizes (La, Lc), No. of layers in aromatic Lamellae have been established as structural parameters for evaluating the carbon stacking structure in carbon materials. These parameters can be determined from the X-ray intensities of coal (Takgi *et al.*, 2004; Sonibare *et al.*, 2010). Raman spectral characteristics, mainly those of the G (graphite) and D (defect) bands at ~1585 and ~1350 cm^{-1}, respectively can provide information for evaluating the degree of ordering and crystallinity in carbonaceous materials (Tuinstra and Koenig, 1970; Robertson, 2002). Tuistra and Koenig noted that the ratio between the intensities of the disorder induced D band and the first order G band, I_D/I_G is inversely proportional to the in-plane crystallite size La or grain size of the graphite domain in carbon (Robertson, 2002). A broad band around 1550-1600 cm^{-1} has been reported in Raman spectra of several carbon-based materials and has been associated with amorphous sp^2 bonded forms of carbon. FTIR is a very useful tool in identifying the functional groups and mineral groups in coal and can provide additional insight into the structural study. Scanning electron microscope coupled with Energy dispersive spectrum (SEM-EDS) is an effective tool used to study the surface characterization and quantification of minerals, oxygen and carbon layers in coal and carbon materials (Wu and Steel, 2007). The UV-VIS-NIR spectra give valuable information about the structure of the molecule because the absorption of UV and visible light involves promotion of electron in σ and $\prod$ orbitals from ground state to higher energy state. The presence of chromophores in a molecule is best documented by UV-Visible spectroscopy (Friedel and Carlson, 1972). These spectroscopic and microscopic techniques are useful in studying carbon nano-structure (Alkhatib *et al.*, 2010; Araoyinbo *et al.*, 2010; Baneto *et al.*, 2010; Fekri *et al.*, 2010; Mamba *et al.*, 2010; Mohamed and Kou, 2011; Ngoy *et al.*, 2011; Zambri *et al.*, 2011).

India has one of the largest coal deposits in World and a leading consumer of coal. For effective utilization of these resources, there is need for scientific investigation into its chemical structure which is scarce today. In this study we have studied the structural parameters of three differently ranked Indian coals varying from sub bituminous to high volatile bituminous by X-ray diffraction, Raman, IR, SEM-EDS and UV-VIs-Spectroscopy.

MATERIALS AND METHODS

Samples and sample preparation: Three Indian coals collected from the coal fields namely; Godavari (G), Korba (K) and Damodar (B) were analysed. The samples were ground into powder and analysed for volatile matter, ash, moisture and elemental composition. The ultimate analysis is carried out and are summarized in Table 1.

The samples were de-mineralised before the X-ray diffraction and spectroscopic analysis to avoid the effect of mineral matter on the quantitative analysis. Five gram of the sample was dispersed in 50 mL of concentrated hydrofluoric acid (HF) solution (40%) and the mixture was stirred for 1 h at room temperature (27°C). The coal filtered and washed with distilled water and dried in an

Table 1: Chemical Analysis and Structural Parameters extracted from the curve-fitting of XRD spectra of coal

Sample	d_{002}	fa	I_{26}/I_{20}	Lc	La	C	O (diff)	V.M
B_1	3.60	0.61	2.14	20.66	27.61	71.66	21.37	36.2
B_4	3.61	0.62	2.01	20.16	26.99	72.58	22.71	35.2
K_1	3.51	0.68	2.85	22.64	28.65	75.74	19.01	31.6
K_4	3.54	0.67	2.65	22.94	28.93	74.39	15.20	32.5
G_1	3.34	0.71	3.28	23.93	38.43	78.04	17.61	24.5
G_2	3.36	0.70	3.07	23.73	39.65	77.21	16.64	24.9

B1, B4: Damodar coal leached with 10 and 40% HF, K1, K4: Korba coal leached with 10 and 40% HF, G1 and G4: Godavari coal leached with 10 and 40% HF. d_{002}: Lattice spacing, fa- Aromaticity, I_{26}/I_{20}-rank, Lc: Stacking height, La: Average lateral size, C: Elemental carbon, V.M: Volatile matter and O: Oxygen content in wt.%

oven at ambient temperature. The leached samples are coded as G4, K4 and B4, respectively in the same order. The chemical leaching was also carried out with 20 and 10% HF on all samples separately.

X-ray diffraction analysis: The XRD data collection was performed by a Bruker AXS D8 Advance X-ray powder Diffractometer at the SAIF, CUSAT, Kochi. Powdered samples were scanned from 4-70° in 2θ range with 0.020° step interval and 2 sec $step^{-1}$ counter time. Origin lab-6 software was used for deconvolution of the diffractogram in the 2θ region 12-32°. The broad hump in this region is fitted two Gaussian peaks around 20 and 26°, namely γ- band and ∏-band (d_{002}), respectively. The peak positions, intensities, widths and area are determined. Theoretically, the areas under the γ and ∏-peaks are believed to be equal to the number of aromatic carbon atoms (C_{ar}) and aliphatic carbon atoms (C_{al}), respectively. Therefore, the aromaticity (fa) of coal i.e., the ratio of carbon atoms in aliphatic chains vs aromatic rings, can be defined as:

$$f_a = \frac{C_{ar}}{c_{ar} + C_{al}} = \frac{A_{002}}{A_{002} + A_{\gamma}} \quad (1)$$

where, A is the integrated area under the corresponding peak. Coal rank is determined from the peak intensities at position 20 and 26° by the equation:

$$\text{Coal rank} = \frac{I_{26}}{I_{20}} \quad (2)$$

The Lateral size (L_a) and the stacking height (L_c) of the crystallite are determined using the Scherrer equation:

$$L_a = \frac{1.84\lambda}{B_a \cos\varphi_a} \quad (3)$$

$$L_c = \frac{0.89\lambda}{Bc\ Cos\varphi_c} \quad (4)$$

where, λ is the wavelength of X-ray used, L_c and L_c are the half width of the (100) and (002) peaks and φ_a and φ_c are the corresponding scattering angles (Takgi *et al.*, 2004; Sonibare *et al.*, 2010).

The parallel arrangement of the aromatic lamellae produces the so-called 001 reflection of which the most intense is 002.

Analysis by spectroscopic technique: The FTIR spectrum was recorded by using Shimadzu FTIR-8400 spectrometer in the region 4000-400 cm^{-1}. The Raman spectra were obtained using a Bruker RFS 100/S spectrometer equipped with a Raman accessory. This comprised a Spectron Laser systems SL301 Nd-YAG laser operating at a wavelength of 1064 nm and a Raman sampling compartment incorporating 180 degree optics. The Raman detector was a highly sensitive standard Ge detector and was operated at 22°C. Under these conditions Raman shifts would be observed in the spectral range 3500- 1200 cm^{-1}. A laser power of 200 mW was used. Diffuse reflectance a spectrum of coals in the UV-Visible and near-IR regions is carried out using a Cary 500 spectrometer equipped with a Praying Mantis diffuse reflectance accessory DRA.

SEM-EDS analysis: SEM micrograph of the virgin and residual samples were obtained by Scanning Electron Micrograph (SEM) JEOL model JSM-6390 LV. The Energy Dispersive Spectrometer (EDS) was obtained by JEOL model JED-2300.

RESULTS AND DISCUSSION

X-ray diffraction analysis: Representative X-ray diffraction profiles for the demineralized coals samples which are fitted for two Gaussian peaks for the band around 26° for the coal samples are shown in Fig. 1. The diffraction profiles show the presence of a clear asymmetric band (002) around 26° which suggests the existence of another band (γ) around 20 o has been reported by many other authors was attributed to the presence of saturated structures such as aliphatic side chains which is attached to the edge of the coal crystallites (Takgi *et al*., 2004; Sonibare *et al*., 2010).

The peak positions, intensity, area and Full Width at Half Maxima (FWHM) obtained after curve-fitting of the (002) and γ bands, calculated structural parameters for all the samples are listed in Table 1. The aromaticity (fa) and I_{26}/I_{20} ratios for the samples range from 0.61-0.71 and 2.14-3.07, respectively. The average lateral sizes (La) and average stacking heights (Lc) of the layer structures in coal samples measures using Scherrer equation (Eq. 1-4) range from 27.61- 39.65 A° and 20.66-23.73 A°. The d_{002} value for two samples (B4 and K4) are higher than that for graphite (3.36-3.37 A°), suggesting a low degree of crystalline order in this sample where as the G4 sample is showing more of graphite structure (3.6-3.34 A°).

Spectroscopic characterization of the coal: The Raman spectra of the coal samples in the range 1300-1700 cm^{-1} with λ = 1064 nm are presented in Fig. 2. The spectrum exhibit a peak at ~1585 cm^{-1} and a weak peak at ~1360 cm^{-1} which corresponds to G and D bands of disordered graphite, respectively. The Raman spectrum of a perfect single graphite crystal would give a main first order sharp peak at 1582 cm^{-1} (G band), due to stretching vibrations of sp^2 bonds in the hexagonal aromatic molecules of graphitic carbon (Manoj and Jose, 2011; Manoj and Kunjomana, 2010b). When disorders is introduced, the G peak broadens between 1580 and 1600 cm^{-1} and an additional peak, known as the D peak, appears between 1350 and 1400 cm^{-1} (Robertson, 2002). In the studied sample the G and D band is more broadened in the case of K4 sample than the G4 sample. The full width at half maximum of the G band is found to be ~80 and 30 cm^{-1} for the K4 and G4 sample, respectively which is far greater than that recorded for carbon nanotubes and highly oriented pyrolytic graphite of about 15-23 cm^{-1} (Robertson, 2002). This feature indicates a low degree of crystalline order in the Korba coal (K4) compared to Godavari coal (G4), confirming

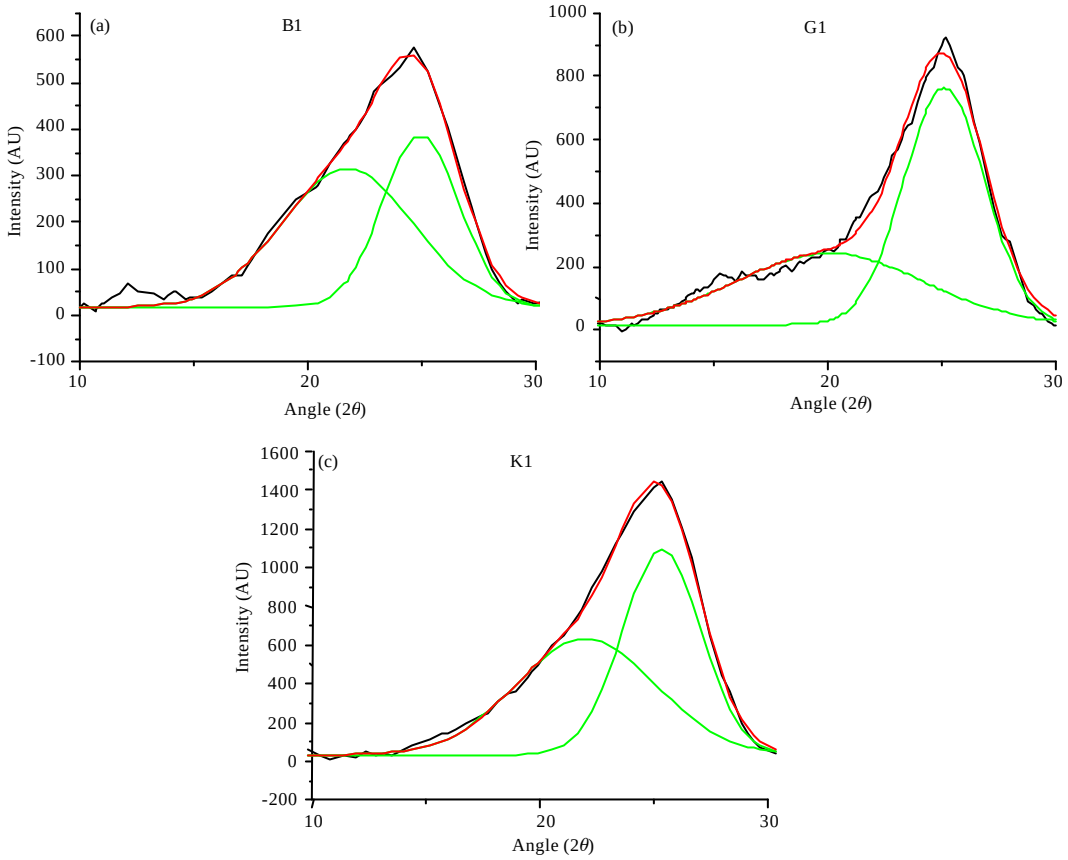

Fig. 1(a-c): Fitting of two Gaussian peaks for the demineralized coal in 2θ~10-30° (green line shows two symmetrical Gaussian peaks fitted which corroborate ∏ and γ band)

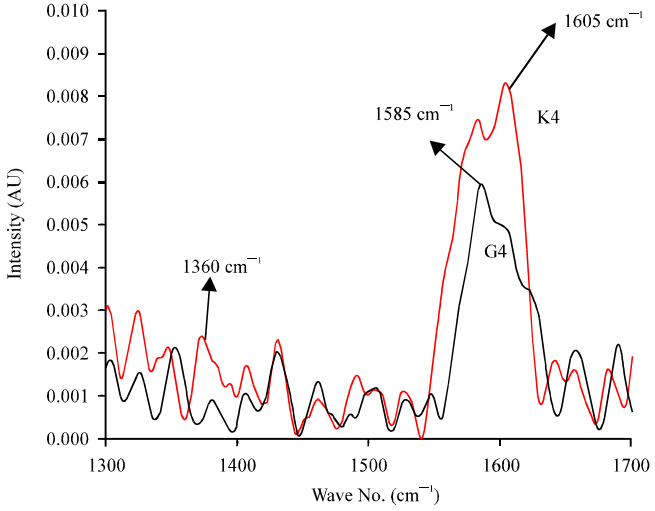

Fig. 2: Raman spectra of demineralized coal samples (KH and GH)

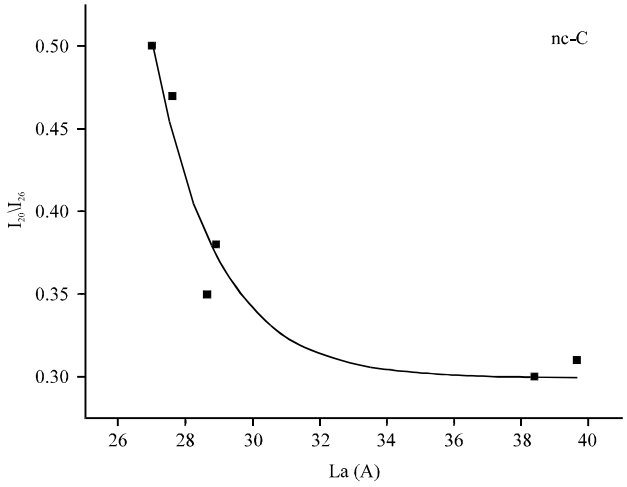

Fig. 3: Variation of I_{20}/I_{26} peak intensity ratio with in-plane correlation length La

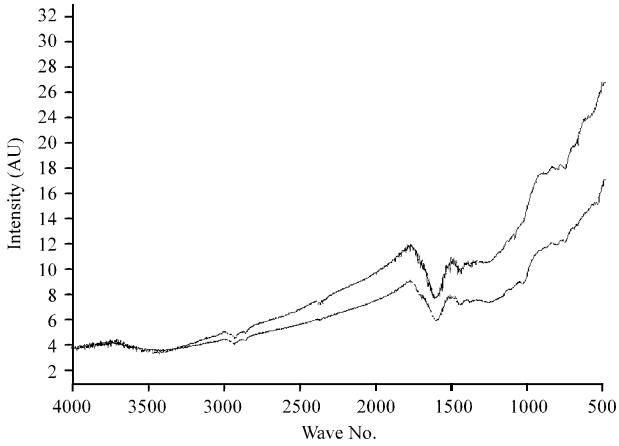

Fig. 4: FTIR spectra of chemical leached coal samples- subbituminous coal (G4) and bituminous coal (K4)

indications from the d_{002} values from the XRD analysis. Tuinstra and Koenig (1970) noted that the intensity ratio of the D and G modes, I_D/I_G, varies inversely with the in-plane correlation length or grain size of the graphite. This equation was based on the Raman spectrum graphite which is highly ordered carbon. In the present study, the intensity ratio I_{20}/I_{26} from X-ray diffraction, is plotted against La values and is shown in Fig. 3. It is found that, the sample is agreeing with Tuistra-Koenig relationship for nano crystalline Carbon. The I_{20}/I_{26} ratio is proportional to the number of rings at the edge of the grain. As the La increases, the intensity decreases quickly.

The FTIR spectra of the demineralized samples are shown in Fig. 4. The coal samples exhibit bands at 2920 and 2850 cm^{-1} due to aliphatic -CH, $-CH_2$ and $-CH_3$ stretching vibration and band around 1450 cm^{-1} assigned to aliphatic -CH bending vibration. The sharp absorption around 1600 cm^{-1} is assigned to the stretching vibration of C = C bonds in aromatic structure. The mineral bands due to Si-O bending vibration occur around 1030-1100 cm^{-1} is very weak in sample due to removal of these minerals with leaching. Low intensity aromatic -CH out-of-plane (bending) bands were observed between 900 and 720 cm^{-1} in all the samples.

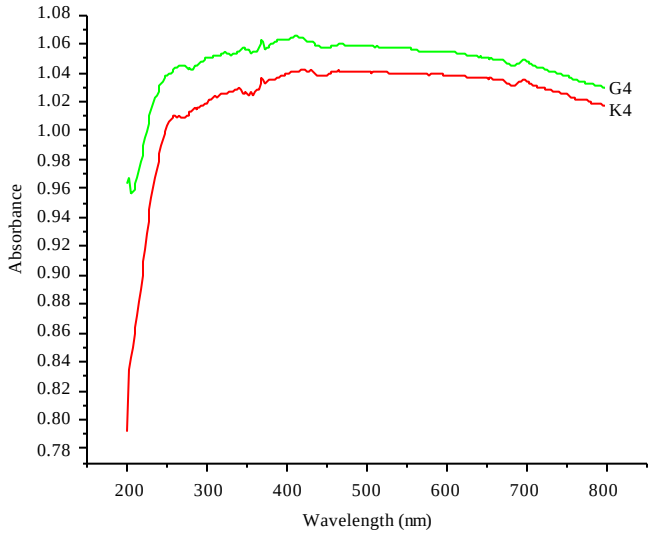

Fig. 5: UV-visible absorption spectra of demineralized coal samples

The electronic absorption spectra of sub-bituminous (G4) and bituminous coal (K4) are measured by the diffuse reflectance spectrometry in the UV-Visible and near IR regions 200-800 nm (Fig. 5). The molecular moieties likely to absorb light in the 200-800 nm region are Pi- electron functions and heteroatoms having non-bonding valence shell electron pairs. Such light absorbing groups are referred to as chromophores. Some chromophores like in C = O, $n\rightarrow\prod^*$ transitions gives rise to λ_{max} at 290 nm and N = O ($n\rightarrow\prod^*$) transition at 275 nm. The general shape of the spectrum is characteristic for hydrocarbons with a single benzene ring. The presence of napthalenoid hydrocarbons will show up in the spectrum and can be judged qualitatively only in the 320 nm regions, where benzoid hydrocarbon give practically no absorption of UV radiation. The two principal bands that are characteristic for the naphthalene system (220 and 280 nm) are masked by the absorption regions of monoaromatic rings; this indicates that the content of napthalenoid hydrocarbon is very low. The sample is almost transparent in the region 475 nm to 650 nm and near IR region. There are small distinct absorption peaks in the UV region and 680 nm. The intensity of the broad absorption in the visible (680 nm) and near IR region (759 nm) are attributable to the $\prod$-$\prod^*$ electronic transitions of the poly nuclear aromatic hydrocarbons which increases with the increase in rank of coal. This is in support to the Raman spectroscopic and X-ray diffraction studies.

SEM-EDS analysis of the sample: The SEM micrograph of the high volatile bituminous coal (K4) and subbituminous coal sample (G4) are presented in Fig. 6 and 7. The surface morphology of the carbon deposit in coal is seen to be non uniform. Large layers carbon sheets are observed on the surface. This layer like structure is due to the formation of graphite layers. Energy Dispersive Spectroscopy (EDS) of the coal shows the presence of carbon and oxygen as main constituent. The composition aggregate from the EDS analysis indicates that studied surface consist of about 98.23% of carbon and 1.77 wt.% of oxygen in the case of sample G4 and 81.1 and 18.85 wt.% in the case of sample K4 with traces of sulphur.

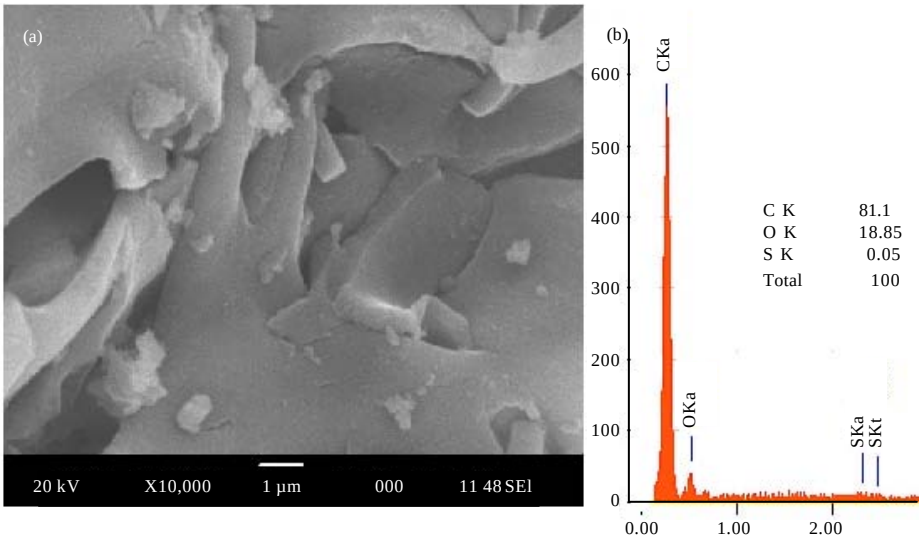

Fig. 6: Observation of surface in bituminous coal (K4) using SEM-EDS

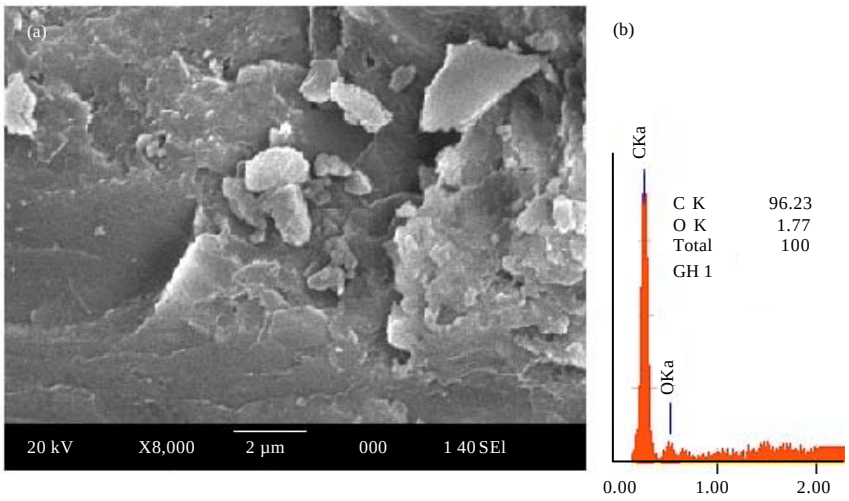

Fig. 7: Observation of surface in subbituminous coal (G4) using SEM-EDS

Relationship between structural parameters and coal rank: The coals samples analysed belong to the sub-bituminous and bituminous group, with the elemental carbon and volatile matter ranging from 71.66-77.21 and 36.20-24.9 wt.%, respectively. In Fig. 8, the aromaticity (fa) values are plotted against I_{26}/I_{20} ratios and it is evident that a high degree of correlation exist between two (0.99). Both parameters measure the degree of maturity in coals. The Elemental carbon contents for the coals are plotted against their fa, d_{002}, rank and Lc values in Fig. 9. There is good correlation between the elemental carbon content and structural parameters (R^2 = 0.97, 0.92, 0.95 and 0.95) of the coal samples. It is known that aromaticity (fa) increases, with decrease in d_{002} as the coal increases in maturity or rank. This observation implies that the structural parameters derived from the XRD studies also reflect the degree of maturity in the studied coal samples, as well as the heat output expected from the coals. The heat output increases with increasing rank. In Fig. 10, the

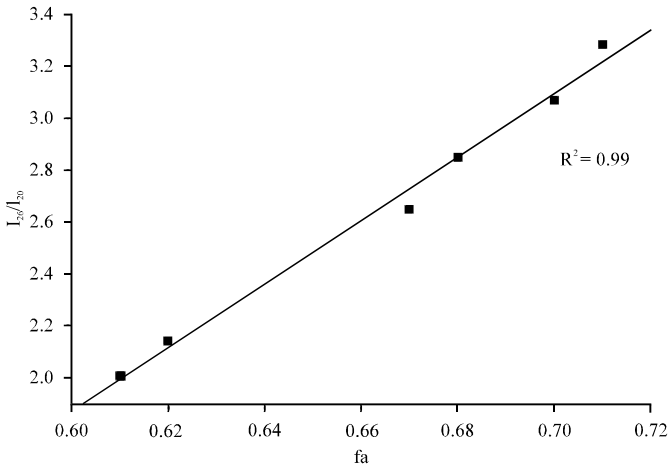

Fig. 8: Relationship between coal structural parameters (aromaticity fa) and I_{26}/I_{20} derived from the X-ray diffraction

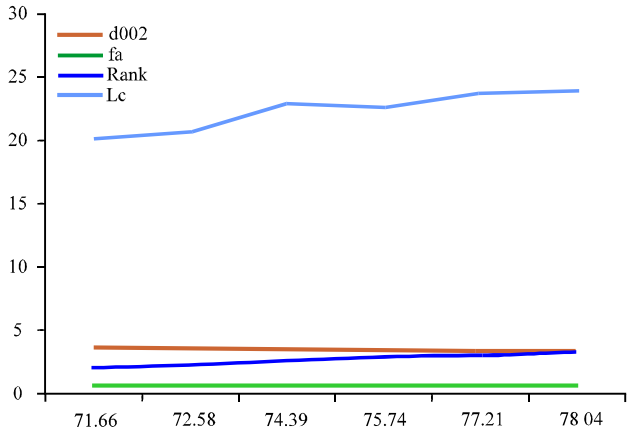

Fig. 9: Relationship between coal elemental carbon and structural parameters (d_{002}, fa, Rank, Lc)

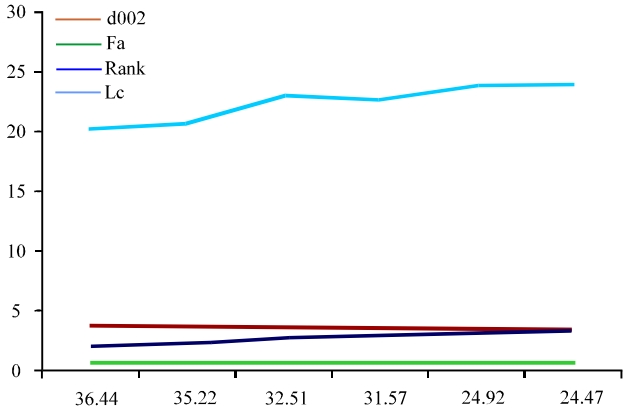

Fig. 10: Relationship between coal volatile matter (VM) and structural parameters (d_{002}, fa, Rank, Lc)

Volatile Matter (VM) values are plotted against the structural parameters derived from the XRD and it is found that a high correlation coefficient exist in al the plots (R^2 = 0.80, 0.89, 0.988 and 0.86), following the same trend as earlier observed for the elemental Carbon content.

CONCLUSIONS

The results of the structural investigation of Indian coal have shown that the coals contain some nano crystalline carbon having turbostratic structure with lateral sizes (La), stacking heights (Lc) and interlayer spacing ranging from 28.15-38.43A° and 22.64-20.16A°, respectively. The Raman spectrum shows broadening with increase of rank. The G band changes from 1585 cm^{-1} to 1605 cm^{-1} for GH sample and KH sample, respectively. This is due to the fact that more disorder is there in the KH sample compared to GH sample. FTIR spectra show that the coal contain aliphatic -CH2 and -CH3, aromatic C = C and -CH bonds in their macromolecular network structures. There are small distinct absorption peaks in the UV region and 680 nm. The intensity of the broad absorption in the visible (680 nm) and near IR region (759 nm) are attributable to the ∏-∏* electronic transitions of the poly nuclear aromatic hydrocarbons which increases with the increase in rank of coal. The SEM analysis shows that the carbon in coal form layered structure. EDS analysis confirms the presence of carbon and oxygen in the surface. A good linear relationship was found to exist between the coal structural parameters and their elemental carbon and volatile matter which reflect their level of maturity. It is observed that the I_{20}/I_{26} ratio is proportional to the number of rings at the edge of the grain. As the La increases, the intensity decreases quickly like in a nano crystalline carbon.

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