Spontaneous combustion of some Permian coal seams of India: An explanation based on microscopic and physico-chemical properties

Rakesh Saxena, G. K. B. Navale, D. Chandra & Y. V. S. Prasad

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Spontaneous combustion of coals is a well known phenomenon in the coal mines of Raniganj and Jharia coalfields. Although several explanations have been given for this burning problem, yet, no general consensus has reached because of variable laboratory techniques. In the present paper, a viable explanation is given for auto-oxidation of coal integrating the microscopic and physico-chemical properties.

Organic petrological investigations of the Permian coal seams reveal vitric and fusovitric petrographic facies corresponding to the Late and Early Permian time, respectively. The characteristic micropetrographic, thermal and chemical properties of the Raniganj coal seams show high to moderate susceptibility to spontaneous combustion. The high amount of the reactive resin/hydrocarbon was found to be closely associated with these coal seams, apart from perhydrous vitrinite having low rank values. In contrast, the Barakar coal seams show least susceptibility probably because of the absence of reactive resin/hydrocarbon with higher rank values.

Microspectrofluorimetric studies suggest green, yellow and orange varieties of reactive resin, hydrocarbons in which green and yellow probably correspond to lipid and terpene fractions of resins imparting more hydroaromatic and unsaturated aromatic structures to these coal in response to their coalification stage. The strong peak at 1600 ± 50 cm in infrared spectrographs of coal also suggests the presence of resin content in Raniganj coals which are rich in C = C, C = O structures. These unsaturated structures play a key role in oxygen-coal reaction leading to the development of an unstable peroxy complex which later through chain mechanism oxidizes the coal while Barakar coals show rigid structural configuration.

The chemical and thermal characteristic *vis a vis.* petrographic composition of these coal seams show conformity in the formation of peroxy complex and its relation with crossing point temperature values. Their crossing point temperature data also suggest conformity in trends with quantitative distribution of the resin/hydrocarbon contents on mmf basis. The endothermic and exothermic peaks and their break down steps revealed by differential thermal analysis and derivative thermogravimetric analysis indicate close correspondence with the coal seams of Raniganj Formation

Thus, it is presumed that the reactive resin content and expulged hydrocarbons from perhydrous vitrinite and organic matrix filled along the cracks, seem to be the main reactive sites for oxygen reaction in spontaneous combustion.

Key-words—Petrology, Fluorescing Vitrinite, Reactive Resin. Peroxy Complex, Raniganj Formation, Barakar Formation (India)

Rakesh Saxena & G. K. B. Navale, Birbal Sahni Institute of Palaeobotany, 53 University Road, Lucknow 226 007, India.

D. Chandra, Department of Applied Geology, Indian School of Mines, Dhanbad 826 004, India.

Y V S. Prasad, Central Coalfield Limited, Darbhanga House, Ranchi 834 008, India.

साराँश

भारत की कुछ परमी युगीन कोयला सीमों का स्वतः दहन : सूक्ष्मदर्शिक एवं भौतिक-रासायनिक गुणों पर आधारित व्याख्या

राकेश सक्सेना, गरुड़ कृष्ण बिन्दिग नवले, डी० चन्द्रा एवं वाई० बी० एँस० प्रसाद

रानीगंज एवं झरिया कोयला-क्षेत्रों की कोयला खानों में कोयलों का स्वतः दहन एक सुविदित घटना है। यद्यपि इसके लिए कई व्याख्यायें दी गई हैं, परन्तु अभी भी कोई सामान्य विचार, जो सभी को मान्य हो, नहीं उभर कर आया है। ऐसा सम्भवतः प्रयोगशाला में विभिन्न विधियों के कारण है। प्रस्तुत शोध-पत्र में सूक्ष्मदर्शीय एवं भौतिक-रासायनिक गुणों के आधार पर कोयले के स्वतःदहन पर स्वयं-शोधित व्याख्या दी गई है।

परमी कोयला-सीमों के कार्बनिक शैलिकीय अन्वेषण से क्रमशः अर्नातम एवं प्रारम्भिक काल की विट्रीय एवं फ़्यूज़ोविट्रीय संलक्षणी व्यक्त होती हैं। रानीगंज कोयला-सीमों के सूक्ष्मशैलिकीय, तापीय एवं रासायनिक गुण स्वतः दहन हेतु उच्च से सामान्य संवेदनशीलता प्रदर्शित करते हैं। क्रियाशील परहाइड्रस विट्रीनाइट की कम मात्रा के अलावा रेज़िन/हाइड्रोकार्बन की अधिक मात्रा भी इन कोयला सीमों में मिलती है। इसके विपरीत बराकार कोयला-सीमों में स्वतः दहन के लिए कम संवेदनशीलता है क्योंकि इनमें क्रियाशील रेज़िन/हाइड्रोकार्बन अनुपस्थित हैं।

सूक्ष्मवर्णफ्लोरोमितीय अध्ययन से क्रियाशील रेज़िन/हाइड्रोकार्बन की हरी, पीली एवं नारंगी किस्में प्रदर्शित होती हैं जिनमें से हरी एवं पीली सम्भवतया लिपिड एवं तारपीन के अंश हैं और ये ही कोयला बनने के समय इन कोयलों में हाइड्रोएरोमेटिक एवं असंतृप्त एरोमेटिक संरचनाओं का योगदान करते हैं। कोयले के निम्न स्पेक्ट्रोग्राफ़ में 1600 ± 50 सेन्टीमीटर पर उच्च अभिलेख रानीगंज के कोयलों में रेज़िन की उपलब्धता इंगित करता है। ये कोयले कार्बन = कार्बन, कार्बन = ऑक्सीजन संरचनाओं से भरपूर हैं। इन असंतृप्त संरचनाओं की ऑक्सीजन-कोयले की क्रिया में महत्वपूर्ण भूमिका है जिसके फलस्वरूप एक अस्थायी परऑक्सी-कॉम्पलेक्स बनता है तथा यही बाद में अन्य श्रृंखलाबद्ध क्रियाओं के माध्यम से कोयले को ऑक्सीकृत कर देता है, जबकि बराकार कोयलों की दृढ़ संतृप्त संरचनायें हैं।

इन कोयलों के रासायनिक एवं तापीय संलक्षण परऑक्सी-कॉम्पलेक्स के निर्माण एवं ताप क्रॉसिंग बिन्दु से अनुरूपता प्रदर्शित करते हैं इनके ताप क्रॉसिंग बिन्दु के आँकड़े ऍम-ऍम-ऍफ० के आधार पर रेज़िन/हाड्रोकार्बन की मात्रा से भी अनुरूपता व्यक्त करते हैं। अतः यह अनुमान लगाया गया है कि क्रियाशील रेज़िन तथा परहाड्रस विट्रीनाइट एवं कार्बनिक द्रव्य से निकले हाड्रोकार्बन, जो कि दरारों में भरे हैं, स्वतः दहन क्रिया में ऑक्सीजन क्रिया के लिए मुख्य क्रियाशील पदार्थों का कार्य करते हैं।

THE Permian coals in India mostly occur all along the depositional tectonic lineaments occupied by the river systems, viz., Damodar, Son, Mahanadi, Wardha, Godavari and the East coast. These coal deposits form the important reserves of fossil fuels in the country apart from the Tertiary coal and lignite deposits.

Pyrohic nature of the coal is a well known phenomenon. In India, this problem is more apparent in the Jharia and Raniganj coalfields of Damodar Valley Basin apart from other coalfields, viz., Hutar, north and south Karanpura, Talchir, Singrauli and Korba. This auto-oxidation character of coal has attracted much attention all over the globe (Kim, 1977; Mazumdar *et al.*, 1983; Ratanasthein, 1983, 1984; Hill *et al.*, 1985; Schmal, 1986; Schmal *et al.*, 1985; Stach *et al.*, 1975; Meuzelaar *et al.*, 1987; Mikula *et al.*, 1987; Chandra *et al.*, 1983, 1987; Banerjee, 1981). Extensive studies have been made to understand this specific property. An overview of the work and the present experimental studies suggest that the physical factors play a significant role, viz., availability and flow of oxygen, particle size, rank of the coal, changes in moisture contents, geological structures and mining conditions. No general consensus has yet reached on the role of various factors independently or interdependently responsible for the spontaneous combustion, because of the variable laboratory techniques. However, the emanating results have conclusively established that the combustion of coal seam occurs when the quantity of heat released by the oxidation of coal is greater than the quantity of heat carried away during the same period of time. The temperature also rises due to the poor heat conductivity of coals. Notzold (1940) opined the importance of rank in the spontaneous combustion. Jones and Towned (1949) and Mazumdar *et al.* (1983) have sugested that higher the rank of coal, lower is its proneness to spontaneous combustion, and also mentioned the possibility of auto-oxidation variation directly with moisture content and concluded that the moisture rich coals are prone to auto-oxidation. Recently, Benerjee (1981) has very critically analysed this problem from various aspects, particularly through the crossing point temperature analyses and DTA techniques after some modifications. Chandra *et al.* (1987) have also shown the role of peroxy index of coals measured at room temperature and also at elevated temperature serving as better control for classifying the coals according to their proneness to spontaneous heating. Apart from the above physical factors, very little attention was given to the petrographic composition of these coals. Ferrari (1938) was the first to indicate the role of vitrite in the outbreak of fire in Heinrich Robert Mine, Ruhr Coalfield, West Germany. Marevich and Travin (1953) have indicated the various petrographic types and their tendency to spontaneous combustion. Munzner (1972) has indicated the danger of spontaneous combustion due to sulphuric acid formed from the weathering of pyrite contents. Valeeva *et al.* (1976) have also recognized the role of liptinite and fusinite indicating their susceptibility to fire.

But the genetic causative factors responsible for spontaneous combustion are yet unknown (Banerjee, 1981). However, Jones and Townend (1949) have indicated that when coal or dump of coal is exposed to atmosphere, it leads to the formation of loose unstable compound, i.e., peroxy complex which through chain mechanism further decomposes and causes 'auto-oxidation'. Thus, it is the coal oxygen reaction which is the main cause for spontaneous combustion. But, what in coal makes it reactive with oxygen, is still a mark of interrogation.

In order to understand the main genetic causative factors responsible for the proneness of coal seams to spontaneous combustion, the present interdisciplinary approach, i.e., field study, megaand micro-petrographic, chemical, thermal, quantitative and qualitative fluorimetric studies have been carried out on the coal seams of Raniganj and Barakar formations from the Raniganj Coalfield, where the problems of spontaneous combustion are severe since long.

GENERAL GEOLOGY

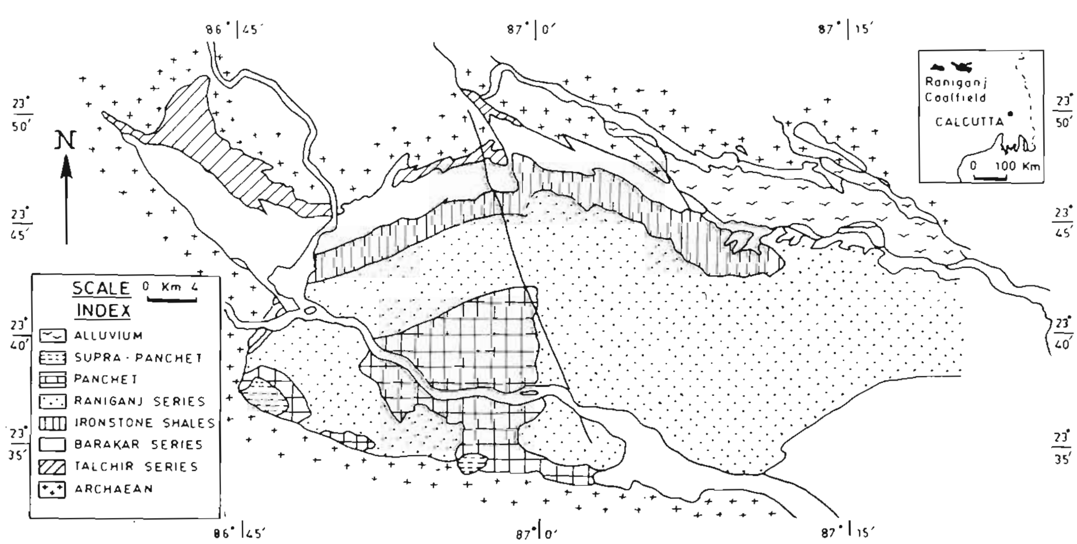
Raniganj Coalfield forms an important coalfield of Damodar Valley from west to East and lies between the longitudes 86 36′ E to 87 20′ E and latitudes 23 35′ N and 23 51′ N It is about 210 km northwest of Calcutta. The coal and associated sediments form two important formations, i.e., Barakar and Raniganj of Early and Late Permian, being the main repository of coals (Table 1).

The Raniganj Coalfield is surrounded by Precambrian rocks on all sides except in the east, where its boundaries are not clear as it is covered by alluvium (Text-fig. 1). In general, the strata trend is almost east-west, in the eastern as well as in the western part of the coalfield. Barakar Formation encompasses seven coal seams regionally which are numbered as B-I to B-VII in ascending order (Table 2). Raniganj Formation comprises 10 coal seams referred to as seams R-I to R-X in ascending order (Table 2).

Samples were collected from the open cast and underground coal seam workings. In all the cases, channel samples were collected from floor to roof as far as possible having time and space relationship. The usual proceedure of coning and quartering was followed to reduce the volume or quantity of each representative sample to 500 gm. Following the standard procedure, representative composite

Table 1—Geological sequence of Lower Gondwana in the Raniganj Coalfield

Age	Formation	Lithological characteristics	Thickness
Tertiary or Lower Jurassic		Dolerite dykes Mica lamprophyre dyke and sills	
Upper Triassic	Supra Panchet	Coarse red, yellow and grey sandstones and quartzose cong lomerates with red shales	305 m
		Unconformity	
Lower Triassic	Panchet Formation	Micaceous grey and yellow sandstones with red clays and green shales	610 m
Upper Permian	Raniganj Formation	Grey and greenish feldspathic sandstones with shales and coal seams	1035 m
Middle Permian	Ironstone shales	Black carbonaceous shales with bands of clay-iron stone	365 m
Lower Permian	Barakar Formation	White and grey coarse to medium grained feldspathic sandstones, shales and coal seams	610 m
Upper Carboniferous	Talchir Formation	Coarse grained sand stones. Fine khaki green shales with boulder bed at the base	275 m
		Unconformity	
Precambrian		Metamorphics	



Text-figure 1—Geological map of Raniganj Coalfield, Bihar, India.

Table 2—Characteristic distribution of various integrated experimental data of the coal seams of Raniganj Coalfield, Bihar

Petrographic & Spectrofluorimetric analysis (Maceral composition under UV light on MMF basis) and Chemical analysis (Air-dried, dmf)

Age	Formation	Coal seam	Range of thickness (in meter)	Frequency of fire accidents in eight years	Maximum reflectance (Ro) microphotometric analysis	Vitrinite: Fluorescing vitrinite	Vitrinite: Nonfluorescing vitrinite	Vitrinite: Total vitrinite	Liptinite: Sporinite	Liptinite: Resinite	Liptinite: Exsudatinite	Liptinite: Liptodetrinite	Liptinite: Total liptinite	Inertinite: Total inertinite	Moisture %	Ash %	Volatile matter %
PERMIAN, UPPER	RANIGANJ	R X	1.20-3.00	32	0.69	50.14	25.96	76.10	12.00	5.00	0.00	3.90	21.00	3.00	4.80	13.40	44.90
		R IX	1.40-6.10	35	0.61	66.25	7.69	73.94	14.25	5.25	—	1.75	21.75	4.00	4.70	8.40	43.55
		R VIII	1.80-13.30	68	0.63	65.23	4.75	69.98	12.52	10.00	-	1.58	24.10	6.00	4.70	9.53	43.76
		R VII	1.80-4.20	30	0.64	69.54	2.98	72.52	8.54	5.00	-	0.46	18.00	10.00	4.50	14.50	41.60
		R VI	1.50-9.00	18	0.71	30.12	39.70	69.82	12.34	3.84	—	1.00	17.18	13.00	3.55	17.35	40.30
		R V	1.20-5.50	25	0.75	21.81	50.24	72.05	9.25	3.01	—	2.00	14.26	14.00	3.60	9.25	37.75
		R IV	2.40-5.80	10	0.73	51.89	13.07	64.96	12.45	3.00	0.84	1.00	17.34	18.00	3.65	9.60	40.05
		R III	1.80-4.50	2	0.74	62.46	6.68	69.14	8.28	8.00	—	1.72	18.00	13.00	3.85	15.45	40.30
		R II	1.20-5.50	3	0.82	49.50	17.50	67.00	12.15	7.43	—	1.00	20.58	12.00	3.60	10.70	37.50
		R I	1.20-1.80	-	0.84	7.62	6.97	64.59	16.43	1.57	-	1.00	19.00	17.00	3.10	15.50	37.80
PERMIAN, LOWER	BARAKAR	B VI	3.00-4.20		1.05	18.36	49.90	68.26	12.56	-	-	2.67	14.23	17.52	2.10	34.40	34.40
		B V	3.00-27.40		0.96	10.33	55.29	65.62	6.42	-	0.50	1.50	8.42	26.00	2.40	33.80	34.10
		B IV	3.00-9.00		0.98	8.89	57.59	66.48	8.46	-	0.05	4.05	12.56	20.96	1.70	30.85	30.85
		B III	7.00-9.00		1.05	14.53	60.41	74.84	6.49	-	-	1.76	8.25	16.91	1.80	17.20	32.80
		B II	2.10-30.00		1.16	8.00	43.52	48.00	8.24	-	-	2.34	10.58	41.42	1.36	27.90	29.10
		B I	1.80-6.40		1.23	10.48	46.52	56.00	8.50	-	—	0.56	9.00	35.00	1.90	15.60	25.90

Thermal analysis

Coal seam	Peroxy complex at room temp. in ($x10^{-5}$ gm equivalent)	Crossing point temperature in °C	Modified peroxy index	DTA Peak intensity (S: Strong, M: Medium, B: Bulge): Endothermic combustion	DTA Peak intensity: Exothermic primary volatilisation	DTG: Decomposition steps in representative samples (°C)	Comparison: DTA-peak temperature in °C (Endothermic)	Comparison: Crossing point in °C
R X	1.26	126.00	1.02	120 S	465 M	120 465	120	126
R IX	4.89	133.50	2.24	120 S	440 M		120	132
R VIII	7.75	135.30	4.28	120 S	465 M		120	136
R VII	8.57	139.00	3.06	130 S	440 M		130	139
R VI	4.69	147.50	1.75	110 S	450 M		130	144
R V	3.61	150.00	2.65	130 S	450 M		130	149
R IV	4.28	147.00	2.04	135 S	450 M	130 450	135	135
R III	6.53	140.60	2.44	135 S	465 M		135	142
R II	5.30	148.00	1.22	150 S	440 M		150	143
R I	3.26	152.00	1.40	175 S	465 M	175 465	175	162
B VI	1.83	166.00	0.61	150 M	465 M	150 460	175	166
B V	2.85	163.00	0.41	150 M	465 M		150	155
B IV	1.01	170.00	0.19	150 M	465 M		150	172
B III	2.86	164.00	0.25	170 B	490 M		170	164
B II	1.63	175.00	0.41	175 B	490 M		175	174
B I	1.43	173.00	0.34	175 B	470 M	175 470	175	173

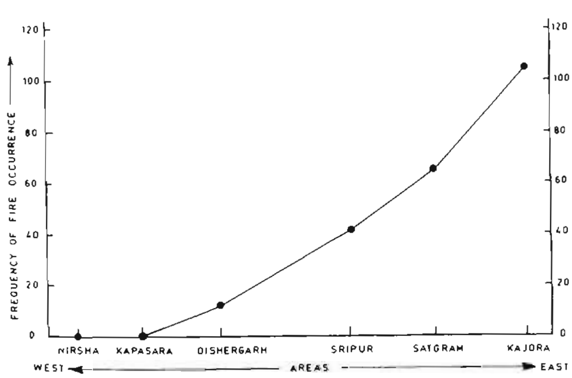
Infra Red analysis, coal facies and fire susceptible zone

Seams	Broad peak at 3500 cm^{-1}	Intense peak 3000 ± 10 cm^{-1}	Two shape and small peak 2800 ± 10 cm^{-1}	Intense peak at 1600 ± 10 cm^{-1}	Peak at 1650 ± 10 cm^{-1} to 1700 ± 10 cm^{-1}	Peak at 790 ± 10 cm^{-1} and at 900 ± 10 cm^{-1}	1080 cm^{-1}, 1010 cm^{-1}, 530 cm^{-1} and 460 cm^{-1}	Coal facies and palaeoclimate	Fire susceptible zone
Raniganj (upper seams)	PROMINENT	HIGHLY INTENSE; UNSATURATED CH-AROMATIC TENDENCY	LESS STRONG; UNSATURATED STRUCTURES IN MOBILE PHASE WITH CH-AROMATIC AND HYDRO AROMATIC TENDENCY WITH π ELECTRON	INTENSE; Resin character tendency with highly unsaturated structures rich in π electron system.	NOT VISIBLE; RESIN CHARACTER	GRADUALLY INCREASE	PROMINENT	VITRIC; REDUCING CONDITIONS IN THE BASIN, PEAT ACCUMULATION KEPT ABREST OF WATER TABLE VERY SLOW WATER FLOW IN SWAMP	UPPER HORIZON; HIGH
Raniganj (lower seams)	PROMINENT	MEDIUM INTENSE; UNSATURATED CH-AROMATIC TENDENCY	MODERATE STRONG; UNSATURATED STRUCTURES IN MOBILE PHASE WITH CH-AROMATIC AND HYDRO AROMATIC TENDENCY WITH π ELECTRON	MODERATE; Resin character tendency with highly unsaturated structures rich in π electron system.	LESS PROMINENT; RESIN CHARACTER	GRADUALLY INCREASE	PROMINENT	VITRIC; REDUCING CONDITIONS IN THE BASIN, PEAT ACCUMULATION KEPT ABREST OF WATER TABLE VERY SLOW WATER FLOW IN SWAMP	MIDDLE HORIZON; MEDIUM
Barakar	LESS PROMINENT	LOW INTENSE	STRONG	Less intense with at shift towards higher wavelength. Polymerising tendency of resin, poor in π electrons	PROMINENT; Condensed structures, i.e. oxidized resin	WEAK	PROMINENT	FUSO VITRIC; DRY AND MOIST CONDITIONS WITH SUBSIDENCE IN BASIN, FAVOURABLE HYDROLOGICAL CON	LOWER HORIZON; LOW

samples were collected from 16 coal seams of different stratigraphic horizons of the Raniganj Coalfield. However, the B-VII coal seam of Barakar Formation could not be collected as there was no workable coal seam available in this horizon.

ACCIDENTS OF SPONTANEOUS COMBUSTION IN THE AREA

Historical records and field study of the fire occurrences due to spontaneous combustion reveal that the total number of fires were 233 for a period of eight years. The frequency of fire occurrences has shown a definite pattern with reference to time and space and is maximum in case of R-VIII seam of Raniganj Formation (Table 3; Text-fig. 4). The



Text-figure 2—Trends of fire occurrence in east-west direction in Raniganj Coalfield.

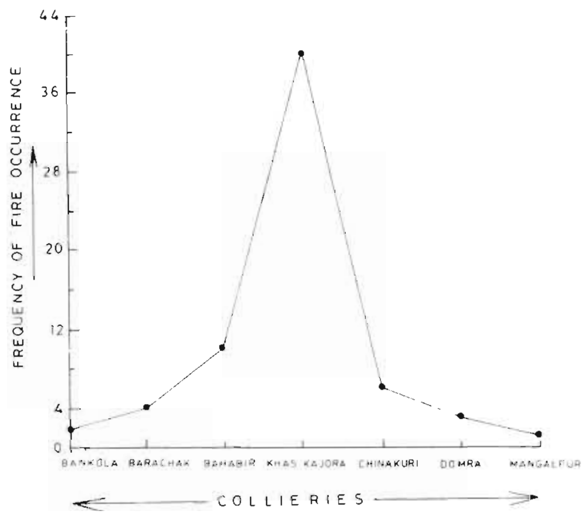
Table 3—Seamwise-yearwise frequency of fire occurrence

Seam Succession	1st	2nd	3rd	4th	5th	6th	7th	8th
R-X	1	4	5	2	10	1	1	8
R-IX	4	3	1	3	2	13	4	5
R-VIII	6	4	5	23	14	9	3	4
R-VII	5	3	5	3	1	2	6	5
R-VI	7	5	2	—	3	—	1	—
R-V	5	3	3	4	4	6	—	—
R-IV	1	1	2	—	2	1	—	3
R-III	—	—	1	—	1	—	—	—
R-II	—	—	1	—	2	—	—	—
R-I	—	—	—	—	—	—	—	—
B-VI								
B-I	—	—	—	—	—	—	—	—

frequency of fire occurrence is more in the upper horizons (R-VII to R-X) and is less in the lower horizons (R-I to R-VI) of the Raniganj Formation whereas no occurrence has been reported in case of Barakar Formation (B-I to B-VII) (Table 3; Text-fig. 4).

PETROGRAPHIC COMPOSITION

Megascopically, the Raniganj coal seams are banded types. The semi-bright and bright bands constitute 50 to 75 per cent of the coal lithotypes. They are normally coarse and thick, varying in thickness from 10 to 38 cm or even more, rendering the coals a friable nature. The bright bands have



Text-figure 3—Occurrence of fire in coal seam-R-VIII in various collieries of Raniganj Coalfield.

sharp contact with dull bands which are sandwitched between the former. The dull bands are thin and consist of fusinite, liptinite and mineral matter. Fusain is occasionally filled with resin/hydrocarbon content of primary and secondary nature It is more abundant in the coals of upper horizon of Raniganj Formation, which are of low rank in comparison to Barakar Formation. This also suggests the potentiality of Raniganj coals to yield more hydrocarbons, i.e., they are more rich in aromatic and hydroaromatic components and have O-rich group, arranged cross wise which causes these coals to have high porosity and a rigid structure (Text-figure 5; Given *et al.*, 1980). Comparatively, the Barakar coal seams are dull banded consisting of high rank vitrinite imparting more condensed aromatic structure in them while inertinite and liptinite groups of macerals and mineral matter form mixed lithologies. Bright layers are thin ranging from few centimeter to 10 cm. The interstitial nature of the mineral composition together with high rank vitrinite has rendered compact nature to the Barakar coal seams.

The bright banded nature of the Raniganj coals has imparted more voids and cleats due to compressional forces for easy access to the expulged fluid to enter, and the air to reach the internal coal surface which is further enhanced by their low rank nature, while in the Barakar coal seams, the access of air to the internal coal surface is very limited because of its compact and high rank nature (Text-fig. 5). However, it does reach the surface, if coal is mylotinized or brecciated.

Micropetrographically, the Raniganj coal seams show characteristic dominance of vitrinitic group of macerals followed by liptinitic and inertinitic maceral groups. Telinite (Pl. 1, figs 3T, 5T) persistently occurs in both Raniganj and Barakar coal seams. The cell lumens are generally filled with reactive resin/hydrocarbons in the Raniganj coal seams (Pl. 1, figs 1R, 2R, 4R, 5R), while in the coal seams of Barakar Formation the collinite (Pl. 1, figs 1c, 2c, 4c) has filled the cell lumen masking the telinitic tissues. Also, the ledges of the epidermal cells are generally filled with micrinite. Absence of micrinite substance in Raniganj coals indirectly suggests that the thermal cracking of liptinite groups of macerals has not reached during the coalification process, where hydrocarbons are released leaving behind the residue of micrinite (Teichmüller, 1987; Khorasani, 1987). While in the Barakar coal seams, micrinite ranges from 1 to 3 per cent only, suggesting the crossing of already expulged hydrocarbon stage. This also confirms from their higher rank values. Collinite (Pl. 1, figs 1-7c) constitutes a major portion of the bright bands and ranges from 39 to 71 per cent in the Raniganj coal seams and 45 to 70 per cent in the Barakar coal seams. They have shown brecciated and cataclastic nature (Pl. 1, fig. 8). Considerable effect of tectonic forces, partial sinking of the basin and displacements of source material have yielded vitrodetrinitic particles (Pl. 2, fig. 3Vt), a characteristic feature of these coals. The common presence of resin duct (Pl. 1, fig. 7R), microcleats (Pl. 1, fig. 8, see arrow) and shear fractures (Pl. 2,

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PLATE 1

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(All the photomicrographs are in oil immersion under normal reflected light)

1. Collinite (c) and Resinite (R) filling in plant tissue.
2. Resinite (R) filling in cell lumen. Groundmass is collinite (c).
3. Telinite (T) compressed tissue, collinite (c) filled and mask the telinite structure.
4. Resin (R) filled in the cell lumen, collinite (c) form the groundmass.
5. Resin (R) filled in cell lumen.
6. Vitrinite along with siderite (Si) mineral matter filling in small cracks.
7. Resin (R) occuring in form of ducts.
8. Cleats developed in vitrite due to impregnation of siderite and other mineral matter

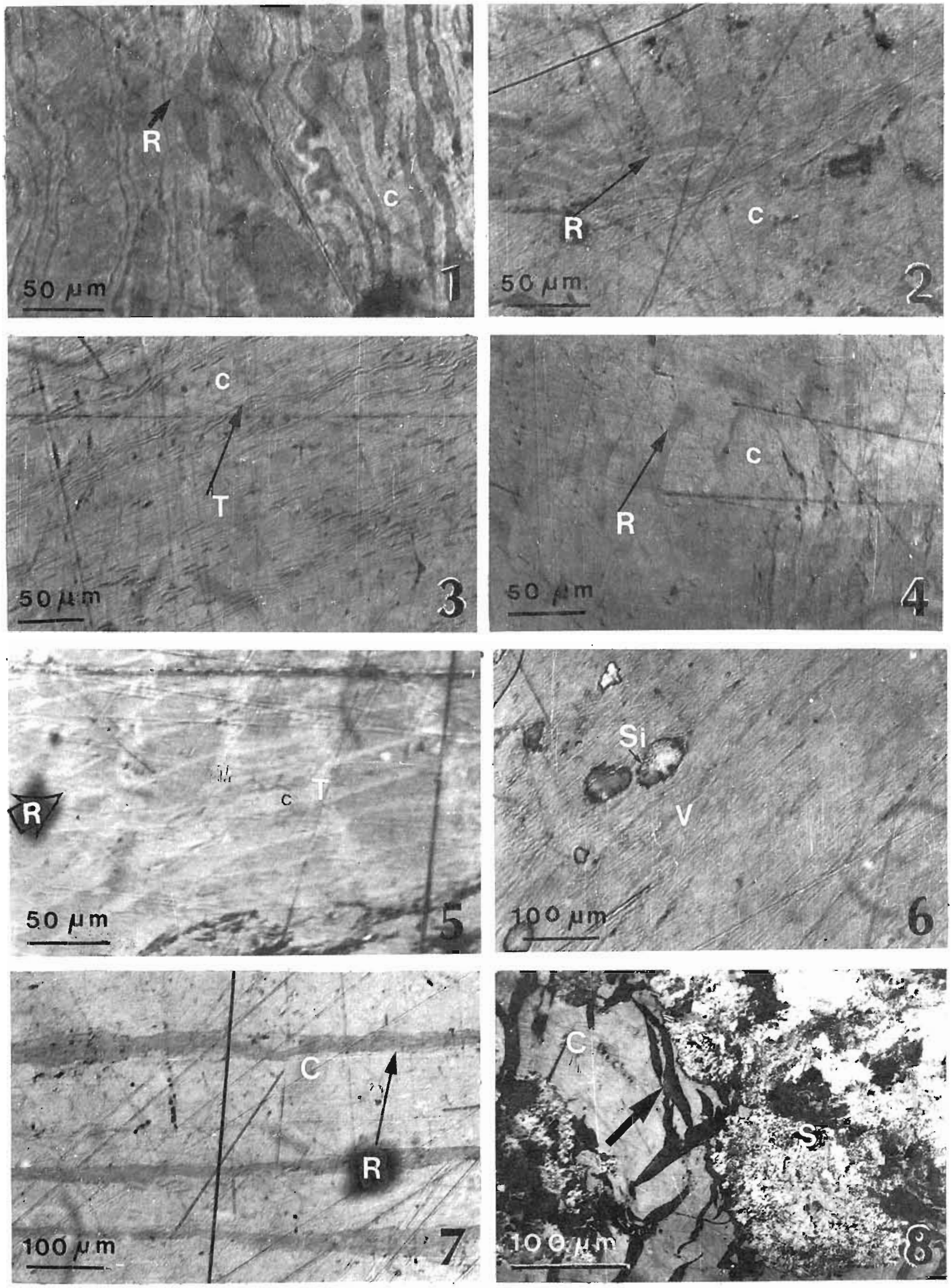
R
c
50 µm
1
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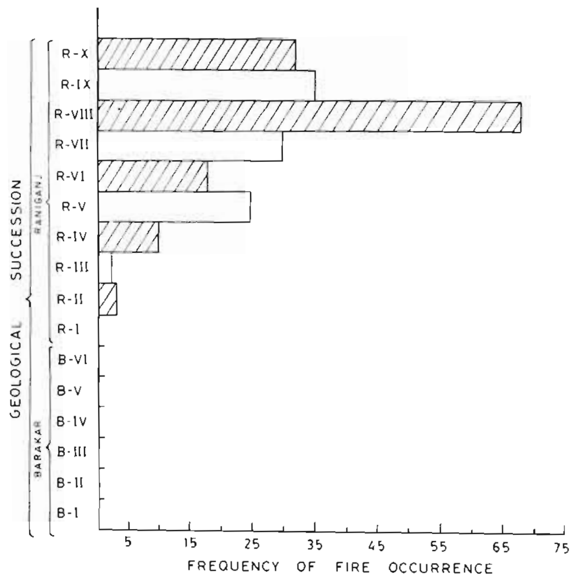
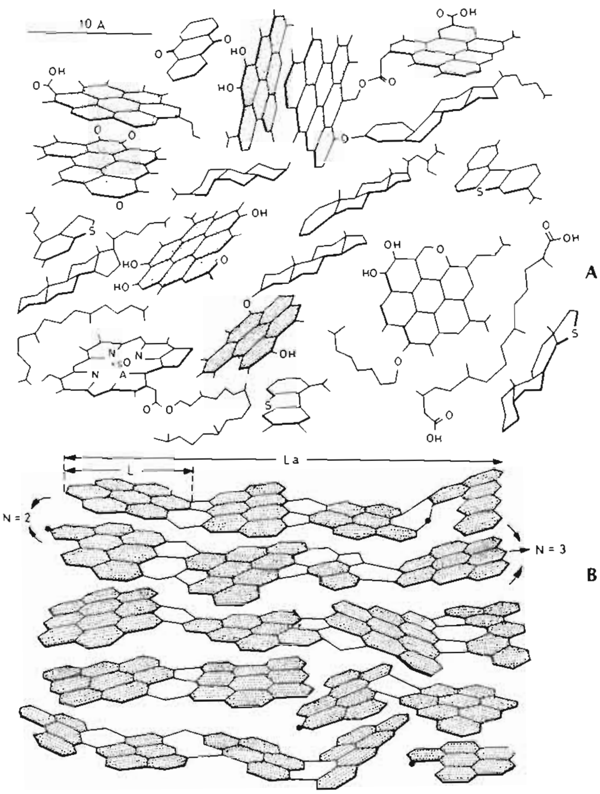
PLATE 1



Text-figure 4—History of fire accidents during a span of 8 years in Raniganj Coalfield.



Text-figure 5—Structure of low (A) and high rank (B) vitrinite (after Oberlin *et al.*, 1980).

fig. 4; Pl. 3, fig. 1) impart secondary porosity to the coals. The microcracks ranging from 3 to 10 μm in width are of three types (i) Parallel to the bedding plane, (ii) right angles to the bedding plane, (iii) irregular cracks (Pl. 2, figs 2, 3, 4, 5; see arrow). These cracks provide easy access of air to the internal surface of the coals and also for the primary and secondary mineral matter impregnation in the Raniganj coals. Further, the microporosity increased in the low rank coals of Raniganj due to more hydroaromatic groups which do not form perfect alignment with the aromatic lamella in the structural configuration of vitrite (Text-fig. 5). The conspicuous occurrence of siderite has been attributed to fresh water conditions of the coal formation (Pl. 5, fig. 1Si). In both, the Raniganj and Barakar coals, the vitrinite forms an important constituent in the composition. However, in the former, liptinite forms the next dominant maceral whereas in the latter, the inertinite group of maceral forms the subdominant entities (Tables 2, 4). The older seams of the Barakar Formation account high inertinite contents over vitrinite particularly in B-I and B-II suggestive of oxidative condition during the formation of the seams. Sporinite forms the characteristic component of dull layers in the Raniganj coal seams and is generally impregnated with reactive resin contents (Pl. 3, figs 2R, 4R; Pl. 4, fig. 4S). Often bituminite specks have also been recorded in these coal seams (Pl. 3, fig. 2B). While, in the Barakar coal seams the sporinite alongwith

PLATE 2

(All the photomicrographs are in oil immersion under normal reflected light)

1. Cataclastic nature of fusinite (F) and vitrodetrinite particles causing high secondary porosity to coal.
2. Compressional irregular cracks in collinite (c), Inertodetrinite (I) in clarodurite.
3. Compressional effect in trimacerite. Vitrodetrinite (Vt) shows brecciated nature, major crack developed in semifusinite (Sf). This provides avenue for free movement of air, oozed out hydrocarbon, resin etc.
4. Microcleats developed in Semifusinite (Sf) showing parallel to bedding plane, right angle to bedding plane and irregular in nature. Maceral contact area between semifusinite and fusinite shows filling of resin (R) content and mineral matter.
5. Trimacerite showing macrinite (Ma), Sporinite (S), Resinite (R) and Inertodetrinite (I).
6. Characteristic preservation of cutinite (Cu) in vitrite band. Cutinite shows ledges (L) in them. Fusinite (F) and macrinite (Ma).

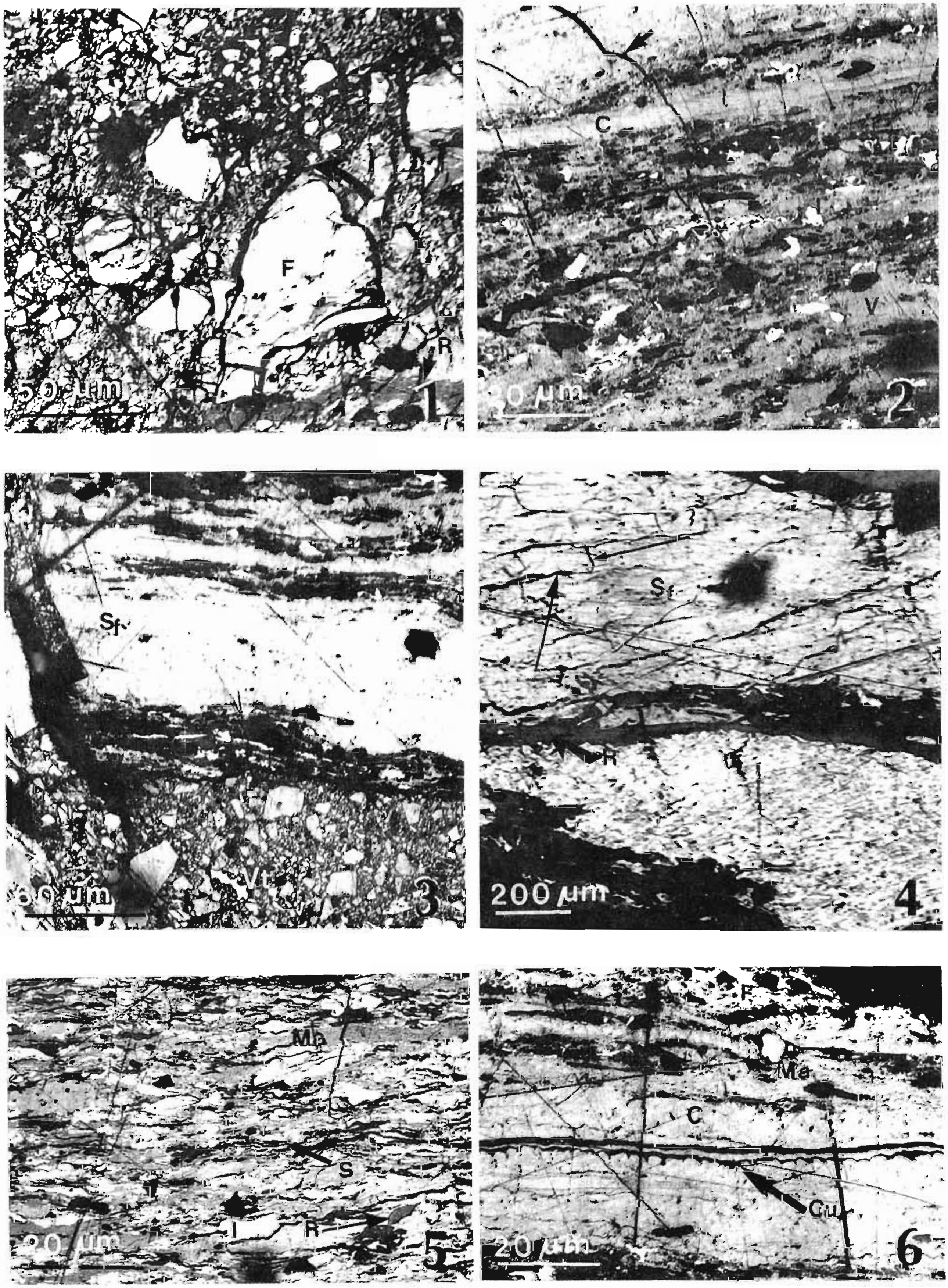


PLATE 2

Table 4—Quantitative distribution of macerals under normal reflected light in Raniganj Coalfield, Bihar

Age	Formation	Coal seams	COAL COMPOSITION						
		MACERAL GROUP	VITRINITE					LIPTINITE	
			Telinite Vit A	Collinite Vit B	Vitrodetrinite	Corpocollinite	Total vitrinite m.m.f.	Sporinite	Resinite
PERMIAN	LATE RANIGANJ	R-X	3.74	70	4.00	—	77.74	16.00	4.54
		R-IX	6.00	65.00	2.48	—	73.48	13.28	4.72
		R-VIII	2.58	65.00	5.00	—	72.58	10.00	9.68
		R-VII	4.00	64.00	2.28	—	70.28	12.00	6.68
		R-VI	3.00	63.00	6.41	—	72.41	14.00	1.21
		R-V	8.71	59.00	4.00	—	71.71	12.0	3.00
		R-IV	4.00	62.00	0.97	—	66.97	16.00	2.00
		R-III	2.03	65.00	—	—	67.03	12.00	5.01
		R-II	2.35	64.00	—	—	66.35	12.00	5.32
		R-I	1.49	64.00	—	—	65.49	15.49	1.00
	EARLY BARAKAR	B-VI	2.10	65.00	3.00		70.10	12.01	—
		B-V	1.23	60.00	3.00	—	64.23	8.05	—
		B-IV	—	58.00	2.80	—	63.80	9.00	—
		B-III	3.31	70.00	—	—	73.40	8.76	—
		B-II	—	45.00	5.23	—	50.23	8.92	—
		B-I	4.54	50.00	3.00	—	57.54	7.89	—

mineral matter content and inertinite, forms clarodurite and duroclarite microlithotypes of dull layers but not filled with reactive resin/hydrocarbon contents. Sporinite forms 10 to 16 per cent in the composition of macerals in the Raniganj coal seams while in Barakar coal seams it ranges from 7 to 12 per cent (Tables 2, 4), under normal reflected light. Cutinite (Pl. 2, fig. 6Cu) is poorly represented being only 1 to 2 per cent in both Raniganj and Barakar coal seams.

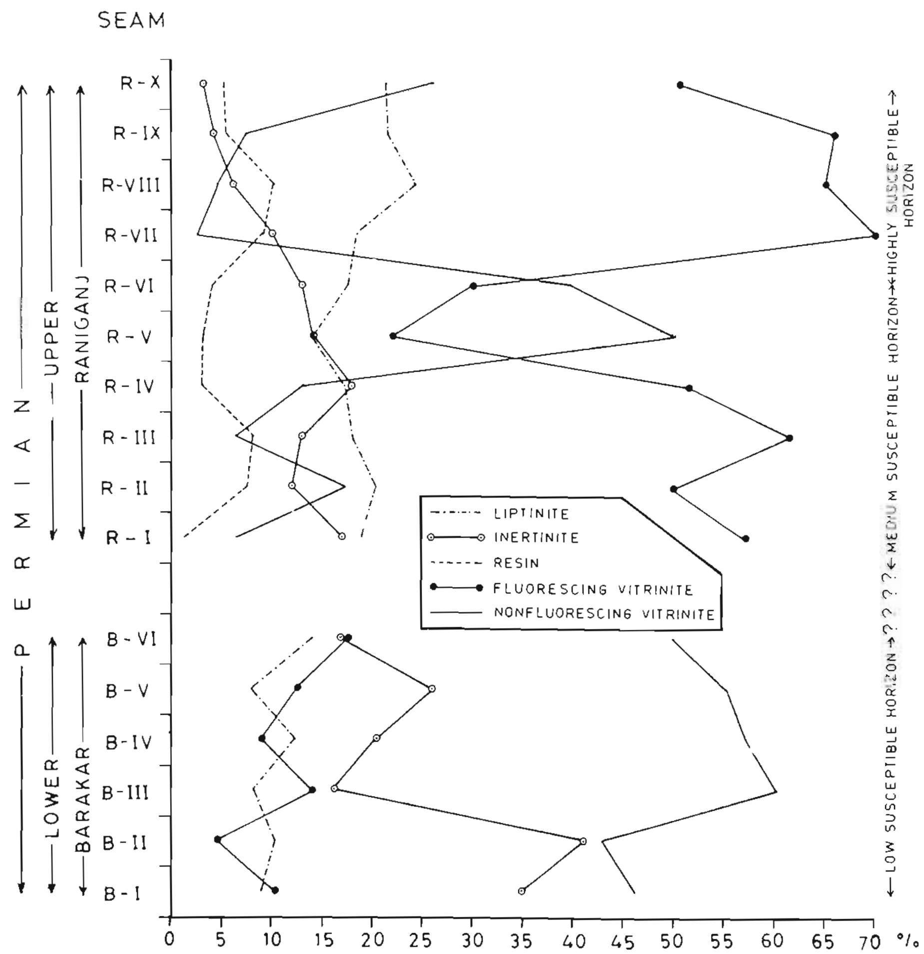
The cutinites in the Raniganj coal seams have shown reactive resinous fillings. Reactive resin/hydrocarbon constitutes a diagnostic entity of the Raniganj coal seams. An increasing tendency in the distribution of the reactive resin/hydrocarbon has been noticed. The older seams show 1.5 per cent while the younger seams account 1 to 9 per cent under normal light (Table 4). It occurs in cell fillings (Pl. 1, figs 1R, 4R, 5R), in the form of ducts (Pl. 1, fig. 7R), in the lumen space (Pl. 3, figs 2R, 4R), in the form of channels (Pl. 3, fig. 1R; Pl. 4, figs 3R, 4R, 5R), alongwith sporinite (Pl. 3, figs 2R, 4R; Pl. 5, fig. 3R), and also along the maceral contact areas, i.e., between fusinite and semifusinite (Pl. 2, fig. 4R). It also occupies the microcrack spaces (Pl. 2, fig. 4R; Pl. 3, fig. 1R) because of its liquid nature. On the basis of the spectral optical properties, the reactive resins/hydrocarbon in the Raniganj coals have been classified in four varieties (Saxena & Navale, MS). The quantitative fluorescence studies have indicated that the reactive resin/hydrocarbon material was not only derived from the liptinitic groups of macerals but also from the perhydrous vitrinitic substance particularly representing the mobile phase of vitrinite network. This mobile phase is rich in polar and CH rich compounds. Yellow and green varieties corresponding probably to terpene and lipid component, respectively were noticed associated with the empty cells of the fusinite (Pl. 6, fig. 6GR), and also oozing out from organic matrix, and vitrite bands (Pl. 6, fig. 4 YR) as recorded by Khorasani (1987) in Australian coals. The lipid rich resin, green in colour under UV light (in blue filter), has also been found in these coals exudating from the liptinitic (bituminite) matrix (Pl. 6, fig. 3GR). A typical greenish-yellow variety of resin/hydrocarbon having lath-shape has been shown (Pl. 6, fig. 9R). The characteristic association of resin/hydrocarbon with the spores (Pl. 6, fig. 10YR) filled along with vitrite cracks (Pl. 6, fig. 5YR) suggests its secondary nature. Liptodetrinite has also been observed in appreciable amount (Pl. 6, fig. 1, Table 2). The remobilized resinite has been observed in the Raniganj coal seams (Pl. 6, fig. 8YR). Occasionally,

		INERTINITE					MINERAL MATTER				
Cutinite	Total Liptinite on m.m.f.	Semifusinite	Fusinite	Inertodetrinite	Micrinite	Total Inertinite on m.m.f. basis	Pyrite	Siderite	Clay	Other mineral matter	Total mineral matter
—	20.54	0.72	1.00	—	—	1.72	1.28	2.00	12.00	7.22	22.50
—	18.00	2.00	6.52	—	—	8.52	1.00	3.00	17.00	2.50	23.50
—	19.68	1.74	6.00	—	—	7.74	1.59	4.00	14.00	2.00	22.25
—	18.68	2.00	8.93	—	—	10.93	1.34	2.00	7.16	3.00	13.50
—	15.21	1.38	10.00	1.00	—	12.38	1.28	3.00	6.00	2.77	13.05
0.52	15.32	1.77	9.00	2.00	—	12.77	1.22	4.00	3.00	1.28	12.50
0.01	18.00	2.00	10.02	3.00	—	15.02	0.36	1.00	6.10	1.29	8.75
—	17.01	1.00	11.38	—	—	15.38	0.54	2.00	6.00	5.00	14.04
—	17.32	4.33	12.00	—	—	16.33	2.43	3.0	7.10	1.72	14.25
—	16.49	4.00	12.00	2.02	—	18.62	1.69	4.00	6.20	3.61	15.50
—	12.01	2.89	9.00	3.00	3.00	17.89	1.56	2.00	17.00	2.94	23.50
—	8.05	1.72	11.50	4.00	0.50	17.72	0.43	4.00	16.00	3.32	23.75
1.32	10.32	0.88	12.00	2.00	1.00	15.88	0.15	6.00	10.0	3.30	20.25
1.76	9.76	1.93	12.18	3.00	0.80	16.93	0.84	3.00	4.21	2.21	10.75
—	8.92	8.00	30.00	1.85	1.00	40.85	0.23	1.00	5.23	1.00	7.50
—	7.89	4.53	24.00	4.00	2.00	34.53	1.64	4.00	6.11	4.00	15.50

inertodetrinite particles of fusinite have been filled with green resin variety (Pl. 6, figs 2I, 2GR). Comparatively, the Barakar coal seams are characterized by oxidized resin bodies (Pl. 5, fig. 6OR) showing distinct resinous nature (Pl. 5, fig. 7OR). Partial oxidized resins (Pl. 5, figs. 6OR, 7OR) have also been noticed in the Raniganj coal seams suggesting fluctuating trends of water level in swamp (oxidative/reducing phases). Charred resin bodies (Pl. 5, fig. 8ChR) have also been recorded from these coals. It has been found that the nature and composition of reactive resins and other macerals are of low rank coal types during the Raniganj period. On the other hand, the Barakar coals lack exsudatinite but well-represented in oxidized resins. The inertinitic group of maceral is meagerly represented in the Raniganj coal seams as compared to the Barakar coal seams. It ranges from 1 to 18 per cent in the former where as in the latter, ranges from 15 to 40 per cent (Tables 2, 4). Semifusinite occurs more in the older coal seams particularly BI, BII of Barakar Formation while in the Raniganj coal seams IX, X and VIII, it is poorly represented. Fusinite has higher frequencies in the Barakar coals ranging from 9 to 30 per cent while in the Raniganj, it ranges from 1 to 12 per cent. The cells and cracks of fusinite are generally impregnated with resin contents and pyritic fractions. Pyrite forms the characteristic association with the younger seams of Raniganj Formation ranging from 0.36 to 2.43 per cent (Table 4), normally disseminated in mineral matter and quite often filled in interstitial spaces of the inertinite macerals. However, in the underlying Barakar coal seams, it ranges from 0.23 to 1.64 per cent. In general, the mineral matter content is more in the coal seams of Barakar Formation.

COAL FACIES

The occurrence, distribution and composition of coal seams reveal that during the Lower Permian coal formation the environmental conditions were oxidative as well as reducing as evidenced by lithology and coal characteristics. The coal seams were mostly dull and of mixed trimacerite type. On the contrary, during the Upper Permian time, the conditions were more suitable for coal formation as shown by extensive thick formation of bright coal bands. The vitrinization and liptinization pathways of coalification were more prevalent in the upper seams indicating vitric facies which suggest enrichment of woody and moist loving plants in the peat swamp (Text-fig. 6). However, due to low



Text-figure 6—Vitrinization, liptinization and inertinization pathways of Permian coal seams of Raniganj Coalfield.

geothermal gradient, the rich vitrinitic coals are of low rank. In the Early Permian time of Barakar Formation, the process of inertinization was more severe and kept pace with the vitrinization path showing vitrofusic facies. It is presumed that the fluctuating trends of water level alongwith the high rate of water flow in the swamp was responsible for inertinite; often, they are filled with mineral matter. Fusinite tissues are hard and seem to have been derived from peat ablation. The characteristic nature of these tissues and their breaking pattern suggests their incorporation at the time of peat deposit derived from forest fire. Few fusinitic tissues reveal secondary porosity in the Raniganj coal seams, due to compressional forces exerted on coal layers as a result of subsidence and also due to the degree of coalification process. This has provided an additional source for the free movement of air, reactive resin/hydrocarbon and mineral matter contents (Pl. 1, fig. 8; Pl. 2, fig. 4R; Pl. 3, figs 1-4; Pl. 4, fig. 2R). The Barakar coals, in contrary, are rich in fusinite, generally filled with mineral matter (Pl. 4, fig. 1; Pl.

PLATE 3

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(All the photomicrographs are in oil immersion under normal reflected light)

1. Trimacerite showing characteristic filling of resin (R) through mega- and microcracks suggesting highly mobile nature. Secondary mineral matter (M) is also filled in certain weak zone.
2. Durite showing semifusinite (Sf), resin (R) fillings, and bitumen (B) filling. Bitumen has been identified under UV light.
3. Clarite showing characteristic cutinite (Cu), sporinite (S), resinite (R) and fusinite (F).
4. Trimacerite showing high incidence of resin (R) fillings in irregular spaces as well as in sporinite, fusinite (F), and vitrinite (V).

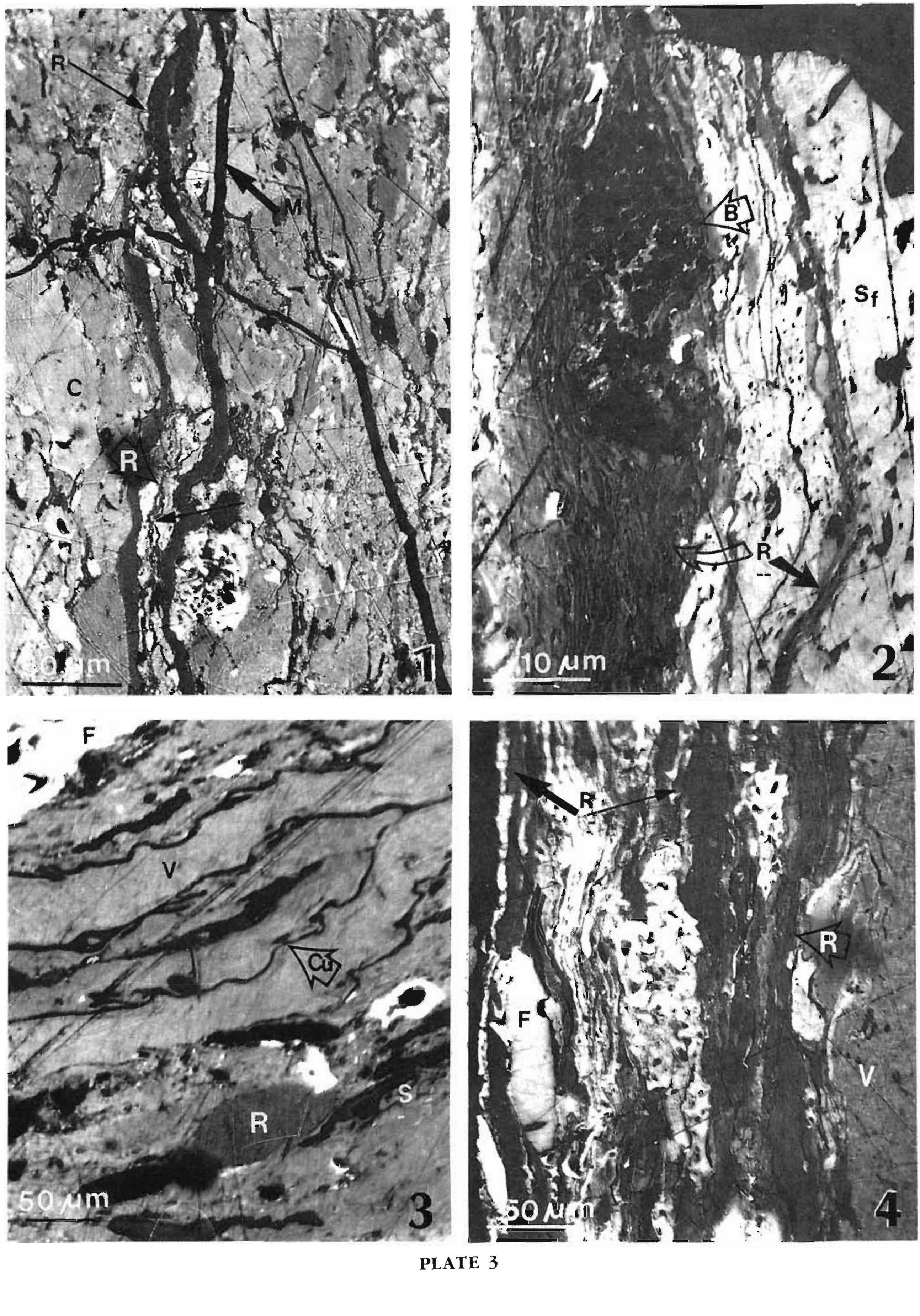
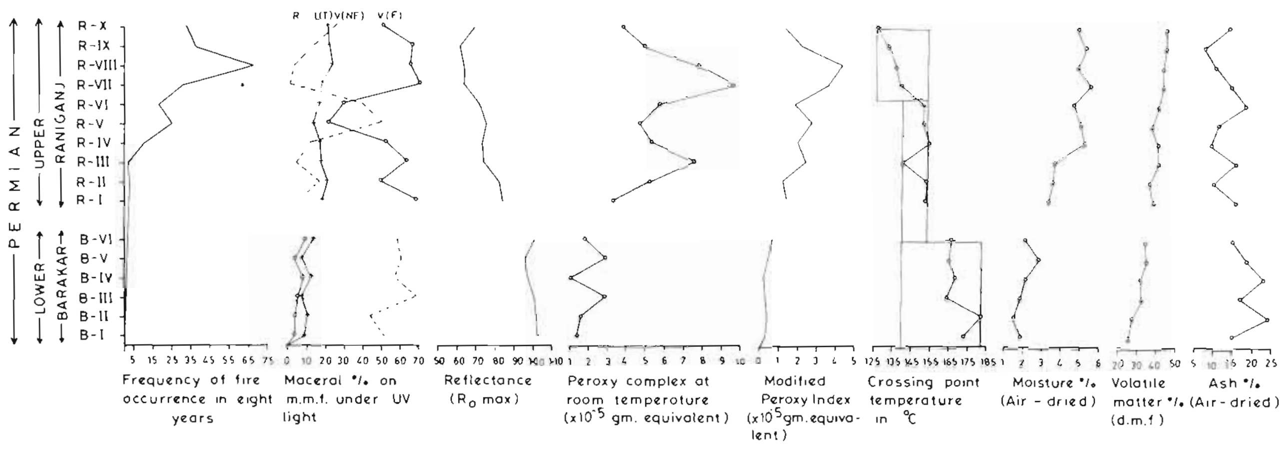


PLATE 3



Text-figure 7—Trends in maceral contents under UV light, reflectance, peroxy complex distribution, modified peroxy index, crossing point temperature, moisture and ash percentage on air dried and volatile matter percentage on dmf in the coal seams of Raniganj Coalfield.

5, fig. 2). These coals generally lack reactive resin/hydrocarbon varieties (yellow/green), however, they are oxidized in nature either due to high rank attained by these seams or due to environmental factors and hence do not show fluorescence properties. This is probably due to highly condensed structural configurations which lost most of their fluorophores (Text-fig. 5). Inertodetrinite occurs persistently both in Raniganj and Barakar coal seams ranging from 1 to 3 per cent and 2 to 4 per cent (Table 4). Micrinite is rare in the seams of Raniganj Formation as compared to Barakar coal seams. This has also suggested the expulsion of resin/hydrocarbon at the preceeding coalification stage from liptinite and vitrinite groups of macerals in the Raniganj coals. Mineral matter present in the coals are mainly clay minerals, pyrite and siderite. Clay mineral (5-17%) occurs in Barakar coals with decreasing tendency through time. While Raniganj coals reveal fluctuating trends. Siderite occurs in all the coal seams uniformly ranging from 2 to 6 per cent. The inter-relationship of vitrinization, inertinization and liptinization paths are quite apparent (Text-fig. 6). Thus, the nature of swamp and palaeoenvironmental factors together with floristic characteristics were responsible for the variable facies development in the seams studied. This is well in conformity with the Permian coal facies recently determined by Navale and Saxena (1989).

RANK

The reflectance of these coals were measured to know the variation of rank in time and space (Table 2). The reflectivity measurements indicated a gradual variation in rank in the lower coal horizons, i.e., coal seams of Barakar Formation (B-I to B-VI) as compared to the upper coal horizons, i.e., coal seams of Raniganj Formation (R-I to R-X). The lower coal seams of Barakar Formation (B-I to B-VI) have shown high reflectivity (Text-fig. 7) which in turn indicates its higher rank, where as the middle

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PLATE 4 →

(All the photomicrographs are in oil immersion under normal reflected light)

1. Fusinite derived from forest fire. Big and small arrows show the breaking patterns and suggest the brittle naure of cell walls which later on got collapsed and filled with resin/mineral matter.
2. Charred woody tissue filled with resin (R) and also shows oxidized corpocollinite structure (Cp).
3. Trimacerite showing filling of resin (R) content in form of elliptical bodies.
4. Trimacerite showing resin (R) fillings, sporinite (S) and also fusinized resin (FR).
5. Clarite showing filling of resin (R) content in elliptical shape. Inertodetrinite (I).
6. Fusinized (F) tissue having oxidized resin globule showing zonning character of resin (R).

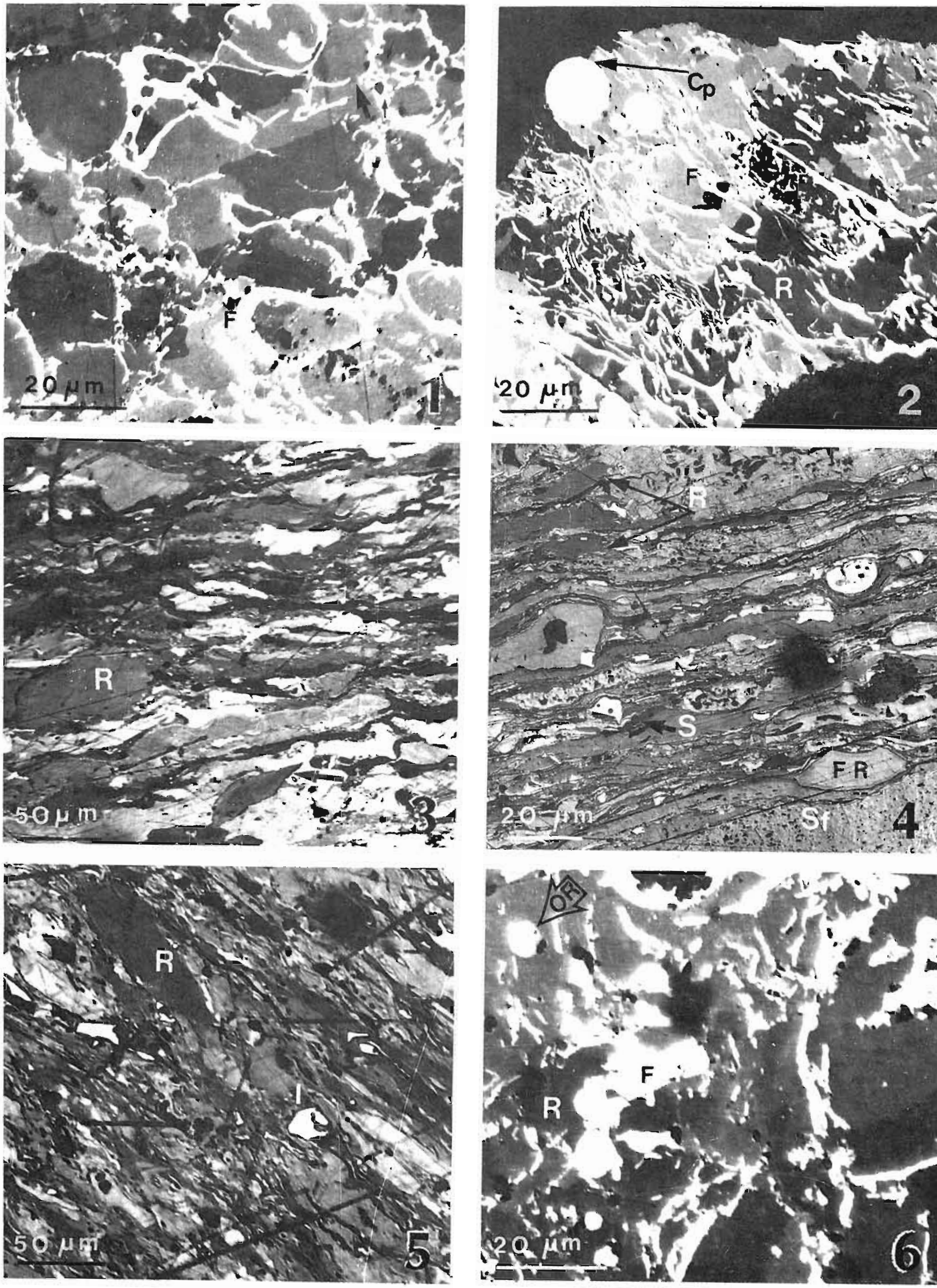
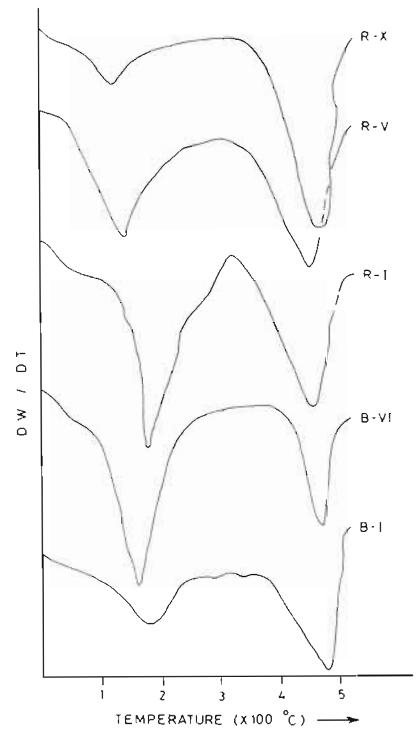


PLATE 4

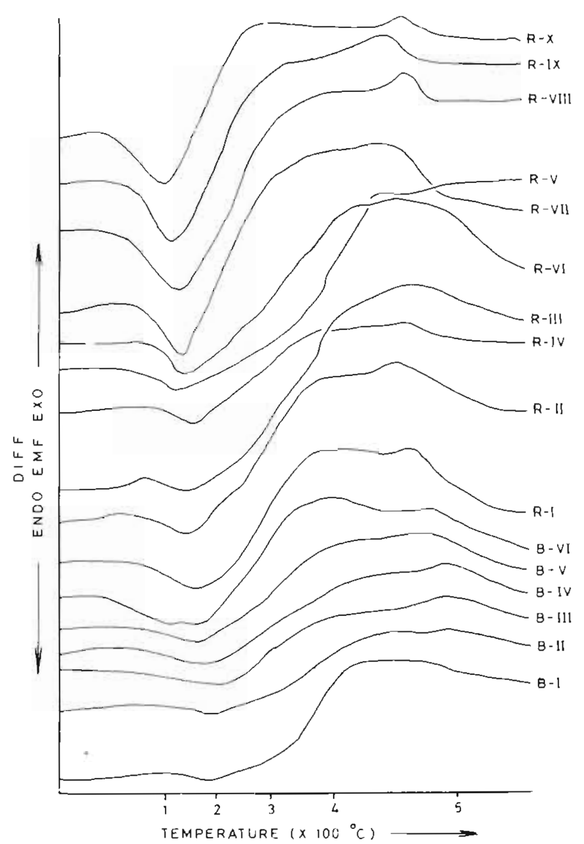
horizons of Raniganj Formation (R-I to R-VI) showed medium reflectivity of these coals. The coal seams of upper coal horizons of Raniganj Formation (R-VII to R-X) showed low reflectivity indicating their low rank. It also suggests the potentiality of Raniganj coals to produce more hydrocarbons, i.e., more rich in aromatic and hydroaromatic component and crosswise arranged O-rich group in macromolecular network and mobile phase of vitrinite structure (Lin *et al.*, 1987; Oberlin *et al.*, 1980). This causes high porosity and rigid structure to these coals (Given *et al.*, 1986).

THERMAL CHARACTERISTICS

The susceptibility of coal to auto-oxidation was known through crossing point method (Ganguly *et al.*, 1953). However, because of several sensitive parameters, viz., flow of oxygen, rate of heating and particle size, etc. any slight change in their values may present erratic results. Further, it is also not possible to simulate the exact mining conditions in the laboratory. Banerjee *et al.* (1967) adopted the DTA technique for understanding the mechanism of spontaneous combustion process. This method was followed in the present investigation to study the thermal behaviour of coal seams.



Text-figure 9—Derivative Thermogravimetric analysis (D.T.G.) of Permian coal seams of Raniganj Coalfield.

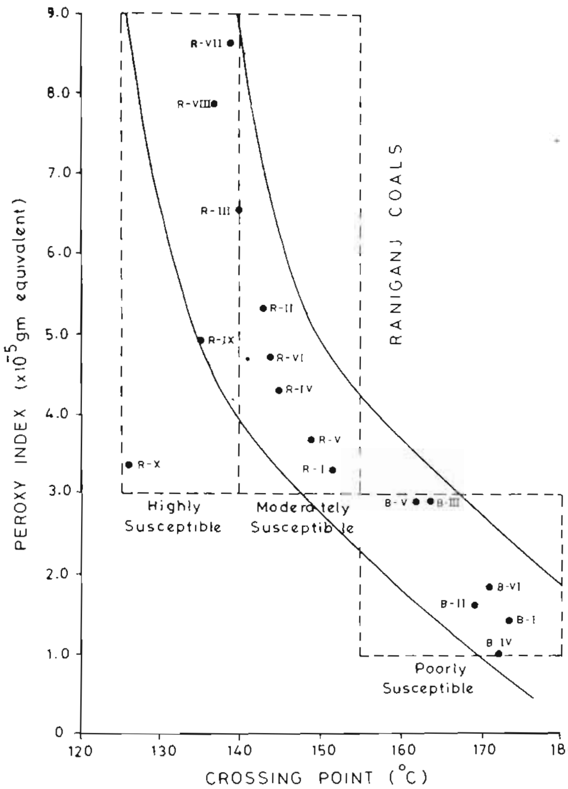


Text-figure 8—Differential thermal analysis (D.T.G.) of the Permian coal seams of Raniganj Coalfield.

The DTA (Differential thermal analysis) curves of the Barakar coal seams showed endothermic bulging peaks in temperature range of 150°-175°C and high temperature exotherms corresponding to 465-490°C (Text-fig. 8; Table 2). The endothermic bulging peaks suggested poor combustion behaviour which was further substantiated by DTG (Drivative thermogravimetry) curves of the representative (B-I to B-VI) samples (Text-fig. 9; Table 2). The breakdown steps of these samples also showed conformity with the above data in the temperature range 150°-175°C and 465°-490°C (Text-fig. 9). The seams of Raniganj Formation (R-I to R-X) showed endothermic peaks in the temperature range from 120°-175°C. The bulging nature of the endothermic peaks showed sharp peak maxima after seam R-III up to the seam R-X. This also shows conformity with the derivative thermogravimetry data of the representative samples (R-I, V, X) of Raniganj Formation (Text-fig. 9). Thus, DTA and DTG curves are in conformity.

Peroxy complex

Self heating characteristics of the Raniganj coals were ascertained from the analysis of peroxy complex formation. This intermediate oxy complex—an unstable compound formed over the coal surface due to the interaction with oxygen of air, is known as peroxy complex. To ascertain the amount of peroxy complex formed by these coals the method of Chalishazar and Spooner (1957) was followed. The contents of peroxy complex at room temperature of different coal seams are tabulated in Table 2 and Text-figure 7. The coal seams of lower horizons, i.e., Barakar Formation (B-I to B-VI) were less reactive in forming the peroxy complex due to their deficiency in reactive resin content and richness in high rank vitrinite which show condensed structural configuration whereas the coal seams of the upper horizons, i.e., Raniganj Formation (R-I to R-X) were more reactive in forming the peroxy complex owing to their richness in high amount of reactive resin/hydrocarbons and perhydrous vitrinite which also contain mobile phase in their structural arrangements. This is also confirmed by their low rank nature (Oberlin *et al.*, 1980). It is in these structures, the oxygen reacts first due to their unsaturated nature and form peroxy complex. This self heating character of these coals has also been further confirmed by the crossing point temperature.



Text-figure 10—Correlation of crossing point temperature (C.P.T.) with peroxy index (P.I.) in Permian coal seams of Raniganj Coalfield.

Crossing Point Temperature

In this method, coal samples were heated in an oxidising atmosphere at a definite rate of temperature rise. The lowest temperature, at which the exothermic reaction in the coal bed can be observed to be self propellant under the experimental conditions, has been termed as the 'crossing point temperature' of the coal concerned. An attempt has been made to classify various Raniganj coals with respect to their susceptibility to spontaneous combustion based on the crossing point temperature (Table 2). Coals which are highly susceptible to spontaneous heating have lower values of crossing point temperature, and poorly susceptible coals have comparatively higher value (Nubling & Warner, 1915). This is probably due to low ignition temperature of reactive resin/hydrocarbon content which quickly helps these coals to reach the ignition temperature where coal catches fire and causes spontaneous combustion. As shown in Text-figure 7, the coal seams of Upper Raniganj Formation (R-VII to R-X) have lower crossing point temperature that ranges from 125° to 140°C whereas the middle horizons of the Raniganj Coalfield, i.e., Lower Raniganj Formation (R-I to R-VI) indicated medium crossing point values ranging from 140° to 155°C, while the coal seams of lower horizons of Barakar Formation (B-I to B-VI) revealed higher crossing point temperature that ranges from 155° to 180°C. This tendency also shows conformity with reactive resins/hydrocarbon content distribution under UV light (Table 2).

The experimental data signified that the coal seams of Barakar Formation (B-I to B-VI) are poorly susceptible to spontaneous combustion, while the coal seams of middle horizons of Raniganj Formation (R-I to R-VI) are moderately susceptible and the upper horizons coal seams (R-VII to R-X) are highly susceptible to spontaneous combustion (Table 2).

As referred by Chandra *et al.* (1983), the susceptibility of coal to spontaneous combustion can be confirmed by correlating the results of crossing point temperature with peroxy index of coal (Text-fig. 10). The relationship suggests that the crossing point temperature increases with the decrease in peroxy index. Coals which are highly susceptible to spontaneous heating would have low crossing point temperature and high peroxy index (also vice versa).

CHEMICAL ANALYSIS

From the chemical analysis, it is apparent that there was a clear cut distinction in chemical behaviour existing in the coal seams of different geological horizons (Table 2).

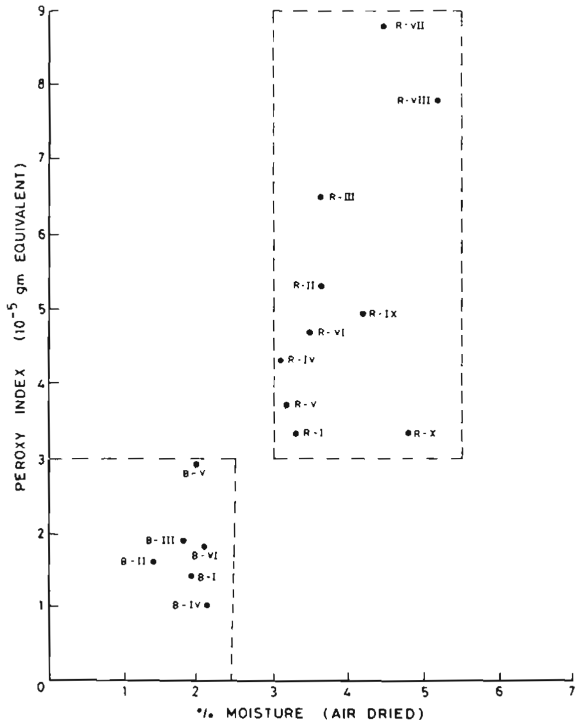
Considerable variation in the moisture was marked in the coal seams of Raniganj Coalfield. In the coal seams of lower horizons (Barakar Formation, B-I to B-VII) the moisture varied from 1.0 to 3.00 per cent. The coal seams of middle horizon (Lower Raniganj Formation, R-I to R-VI) contain the moisture in between 3.00 to 4.00 per cent, and in the seams of upper horizons (Upper Raniganj Formation R-VII to R-X), the moisture content varied from 4.00 to 5.50 per cent (Text-fig. 7). Thus, there is a gradual increase in moisture content from the coal seams of lower horizons to upper horizons.

Significant variation in the percentage of volatile matter on dry mineral matter free basis (dmf) was observed in the coal seams of different horizons. The variation range in moisture and volatile matter in different coal seams is shown in Table 2. It can be seen from Table 5 that the volatile matter percentage of four coal horizons in the Upper Raniganj Formation lies within the narrow range of 41.00 to 45.00 per cent. Whereas the seams of middle horizons (R-I to R-VI) have volatile matter in between 36.00 to 41.00 per cent. The seams of lower horizons of the Raniganj Coalfield (Barakar Formation, B-I to B-VI) showed a wide variation in volatile matter ranging in between 25.00 to 36.00 per cent. However, the ash content was found to be randomly distributed in the coal seams of different horizons (Text-fig. 7). In general, the percentage of moisture and volatile matter in the coal seams gradually increased from lower to upper horizons, and has influenced the susceptibility of these coal seams to spontaneous combustion.

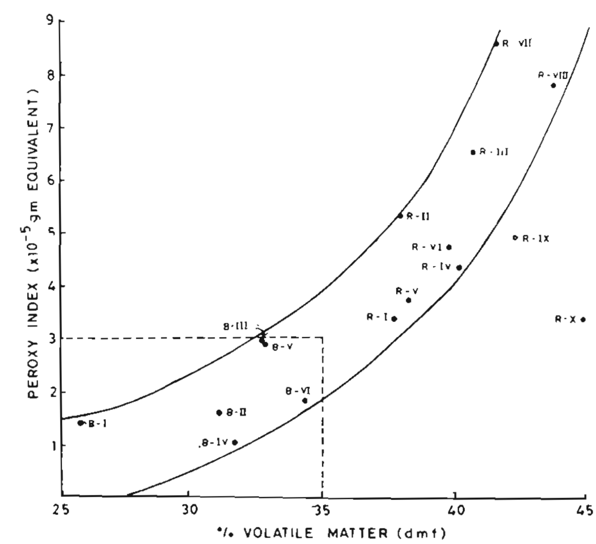
To interpret the relation between the chemical behaviour and the ability of coal to form peroxy complex, the moisture content was plotted against peroxy complex. As shown in Text-figure 11 the high moisture coals of the Raniganj Formation (R-I to R-X) have higher values of peroxy complex content than the lower moisture coals of Barakar Formation (B-I to B-VI). While the high volatile coals of Raniganj Formation (R-I to R-X) were more reactive in providing higher values of peroxy complex (Text-

Table 5—Variation of moisture and volatile matter in the coal seams of the Raniganj Coalfield

Formation	Coal horizon	Range of moisture (Air dried) %	Range of volatile matter (dmf) %
Upper Raniganj Formation	R-VII to R-X	4.00-5.50	41.00-45.00
Lower Raniganj Formation	R-I to R-VI	3.00-4.00	36.00-41.00
Barakar Formation	B-I to B-VI	1.00-3.00	25.00-36.00



Text-figure 11—Correlation of moisture versus peroxy complex (P.O.C.) in Permian coal seams of Raniganj Coalfield.

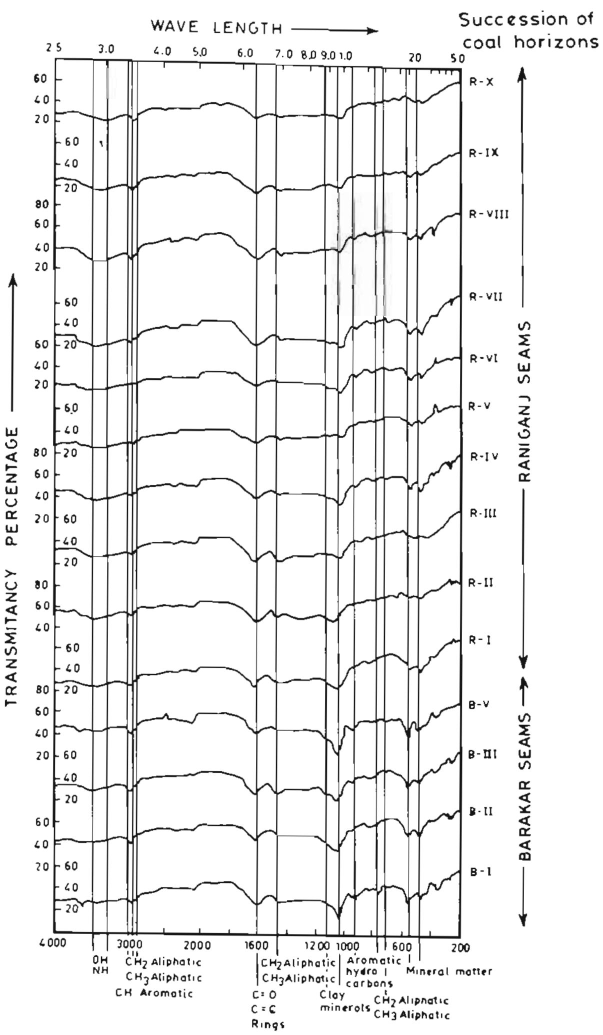


Text-figure 12—Correlation of volatile content and peroxy complex (P.O.C.) distribution in Permian coal seams of Raniganj Coalfield.

fig. 12) than the low volatile coals of Barakar Formation (B-I to B-VI). Thus, the coal seams of lower horizons (B-I to B-VI) are less susceptible to spontaneous combustion than the coal seams of upper horizons (R-I to R-X).

It is further observed that the high moisture coals of the Raniganj Formation (R-I to R-X) showed low crossing point temperature; while the low moisture coals of Barakar Formation (B-I to B-VI) showed high crossing point (Text-fig. 7). In other words, high moisture coals of the Raniganj Formation (R-I to R-X) are more prone to spontaneous combustion than the low moisture coals (B-I to B-VI) of Barakar Formation.

Similarly, high volatile coals of Raniganj Formation (R-I to R-X) have shown low crossing point temperature and the low volatile coals of Barakar Formation (B-I to B-VI) show high crossing point temperature (Text-fig. 7). Thus, the susceptibility to spontaneous combustion varies with moisture and volatile content of coals.



Text-figure 13—Infra-red characteristic of Permian coal seams of Raniganj Coalfield

INFRA-RED STUDIES

The infra-red spectral characteristics from the coal samples of different seams of Raniganj Coalfield (Text-fig. 13) suggested variation in functional groups with reference to spontaneous combustion:

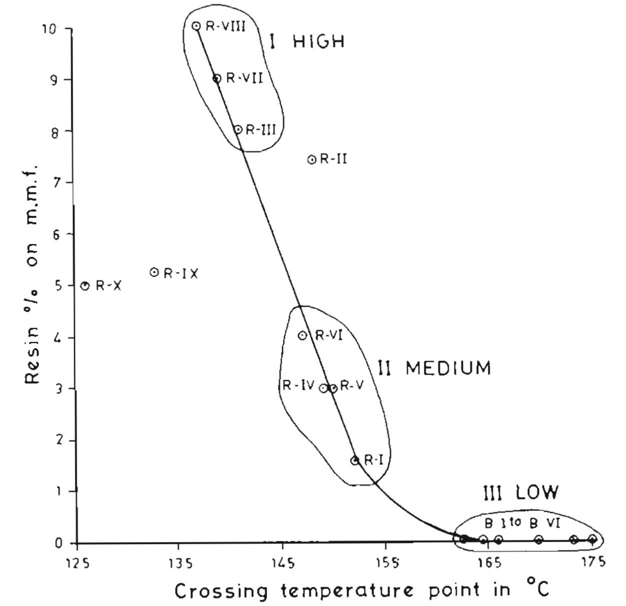
1. An observed broad peak 3000 cm^{-1} to 3700 cm^{-1} undoubtedly represents intermolecular hydrogen bonded OH group in the organic material of the coal. Main peaks are around 3500 cm^{-1} in almost all the samples along with minor peaks at 3250 cm^{-1}. Hydrogen bonded NH group may also contribute partly to the intensity of the band at 3300 cm^{-1}. This broad peak was noticed more prominent in the case of Raniganj seams than the Barakar seams. It indicates a high moisture absorbing nature of the Raniganj coal seams. Nandi *et al.* (1963) had concluded earlier that amenability of a coal to self heating as indicated by lower crossing point temperature was due to the hydroxyl groups of the coal structures.

2. CH-Aromatic stretching frequency was found to be weaker in the coals of lower horizons of Barakar Formation (B-I, II, III, V) at 3000 ± 20 cm^{-1}. While it is with moderate intensity in the middle horizons (R-I to R-VI) and stronger in the upper most horizons of Raniganj Formation (R-VII to R-X) as shown in Text-figure 13. Choudhary *et al.* (1982) established that the higher the aromatic content, the faster is rate of auto-oxidation and lower is the crossing point temperature. Thus, CH-Aromatic content could be playing an active role in the susceptibility to spontaneous combustion of seams of upper horizons (R-VII to R-X).

3. Most of the samples showed characteristic absorption at 2900 ± 10 cm^{-1} and near 2850 ± 10 cm^{-1}. These stretching absorption points have indicated the presence of aliphatic groups. Aliphatic CH and CH groups contributed to the intensity of the bands. These aliphatic absorption bands have been found to be stronger in the seams of lower horizon (Barakar Formation, B-I, II, III, V); moderately intense in the seams of middle horizon (R-I to R-VI) and less intense in the seams of upper horizons (R-VIII to R-X). These aliphatic chains also occurred at 1440 cm^{-1}. The intensitites of these bands were similar in different seams in the same pattern as mentioned above.

4. A very strong absorption peak occurs at 1600 ± 10 cm^{-1} in all the spectra. It is probably due to several groupings as C = C, oxygen containing groups as C = O and hydrogen bonded carboxyl

group. The peaks recorded in the coals are more intense in the Raniganj coal seams. The peaks of seam R-II, R-III, R-VII, R-VIII and R-IX show conformity in their trends with each other. These seams are also characterized by high amount of reactive resin-hydrocarbon contents while the peaks observed in R-I and R-V having similarities with each other in its characteristics, viz., shape, sloping behaviour, etc. have also slightly shifted towards higher wavelength 1650 ± 10 cm^{-1} (seam R-I) than the above younger seams of Raniganj Formation. This also corresponds to the quantitative distribution of green and yellow fluorescing reactive resin hydrocarbon and fluorescing vitrinite contents on m.m.f. basis (Table 2). Further, the seam R-I of Raniganj Formation has higher rank value which also contributes for shifting of peak towards higher wavelength side (Murchison, 1966) The characteristic fluorescing nature of the Raniganj coals suggests the occurrence of various fluorophores containing π electron system in them which has been presumed to emit fluorescence signals and is mainly associated with the unsaturated structures. These unsaturated structures whether as the part of mobile phase of vitrinite (Lin *et al.*, 1987) or of reactive resin/hydrocarbon provide main reactive sites for oxygen coal reaction and results in the development of high amount of peroxy complex. On the contrary, the absorption peaks of the Barakar coal seams show less intense nature having similarities with each other in their characteristics and are apparently different from the Raniganj coals. The peaks have shifted slightly towards the higher wavelength side, viz., seam B-V This is probably due to high rank nature. Spectrofluorometric properties of the Barakar coal seams have shown their deficiency in reactive resin/hydrocarbon content, and amount of fluorescing vitrinite, or in other words, the resin contents of the Barakar coal seams are not reactive and consist of saturated structures, i.e., they do not have fluorophores having π electron system which quenches the flourescing properties. This is corroborated by the occurrence of oxidized resin bodies recorded in Barakar coals and their high rank nature. As the rank increases the organic



Text-figure 14—Relationship of resin content on mmf basis vs crossing point temperature in Raniganj Coalfield.

structures became more polymerized and condensed and quench their fluorescing properties (Lin *et al.*, 1987). It is known that the presence of strong peaks at 1700 cm^{-1}, 1450 cm^{-1} and 1380 cm^{-1} in resin from low rank coal (lignite) gets slightly shifted towards higher wavelength side and has distorted peak characteristic as the rank increased in bituminous coal (Murchison, 1966). Concomitantly, IR spectra of the resin from bituminous coals show intense peak at 1600 cm^{-1}. The peaks in Raniganj coals occur at 1600 cm^{-1} while in Barakar coals, their nature is different and slightly shifted with impaired spectra (B-V). IR spectral properties have been recognized in fusinitized resin rodlets from Pomeroy coal (Upper Pennsylvanian), Holmes Mine, Kanawha County, West Virginia (Lyons *et al.*, 1982). The impared characteristic spectra also suggest the occurrence of high resin contents as well as unsaturated aromatic and hydroaromatic structures in the Raniganj coals in comparison to Barakar coals (Text-fig. 13; Table 2).

Small peaks recorded at 790 ± 10 cm^{-1} and 90 ± 10 cm^{-1} were due to the stretching of aromatic

PLATE 5 →

(All the photomicrographs are in oil immersion under normal reflected light)

1. Siderite (Si) nodules in trimacerite band. Sporinite (S), fusinite (F), tellocollinite (Tc).
2. Fusinized (F) tissue filled with mineral matter.
3. Resin (R) fillings along the cracks, non structured fusinte (F).
4. Partially oxidized resin (POR) in vitrite.
5. Syngenetic mineral matter concretions (Mm) in vitrite band showing pressure direction.
6. Oxidized resin (OR) in trimacerite.
7. Oxidized resin (OR) showing differential zonning character
8. Charred resin body (Ch. R) in vitrite band.

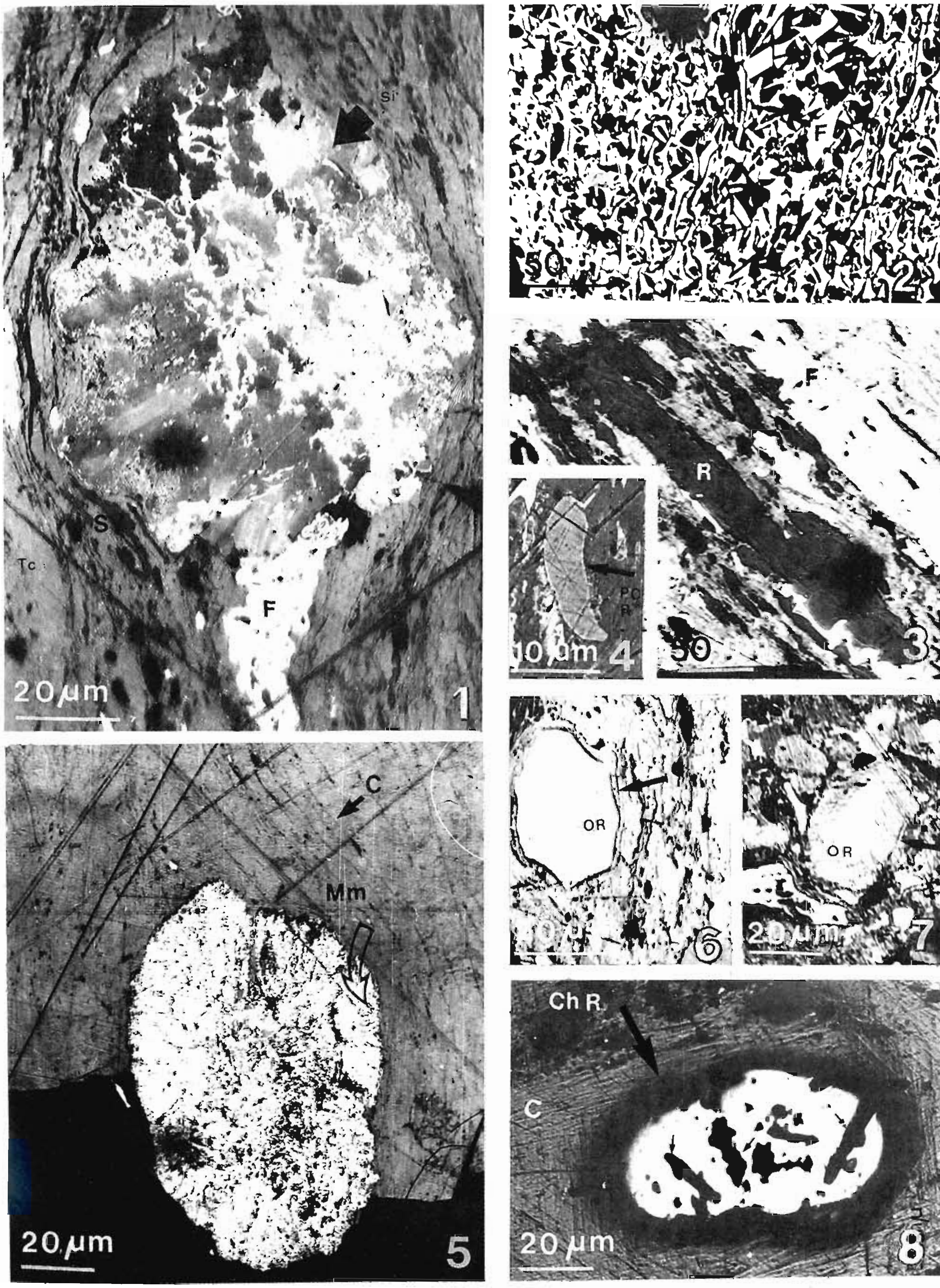
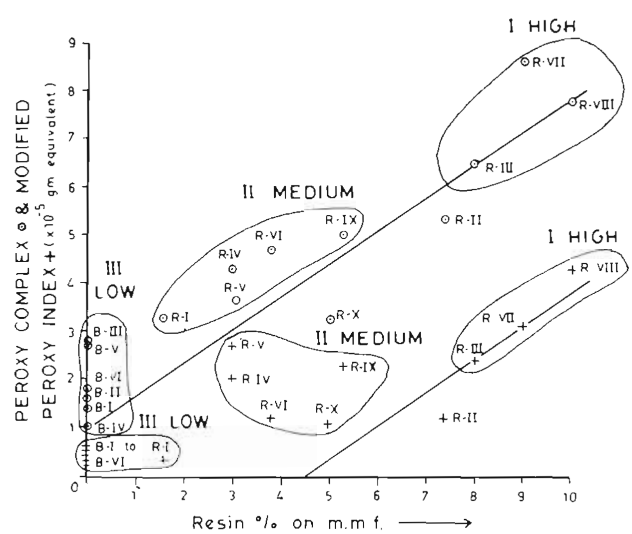

Si
S
Tc
F
20 µm
1
F
50
2
F
R
50
3
PC
R
10 µm
4
C
Mm
20 µm
5
OR
6
OR
20 µm
7
Ch R
C
20 µm
8

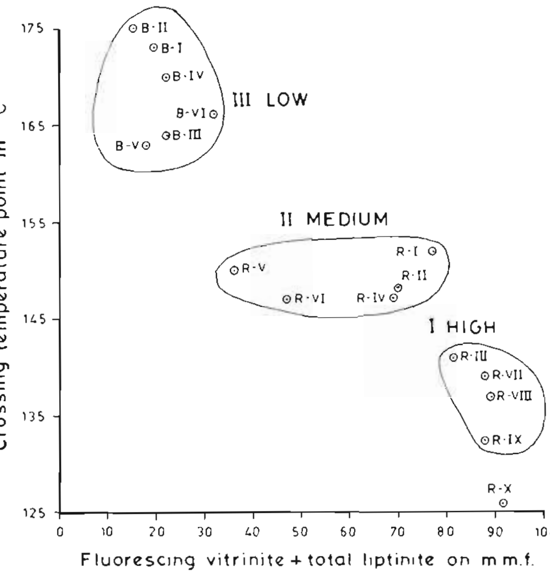
PLATE 5

hydrocarbons. The absorption bands in this region may be associated with aromatic C-H out of plane deformation vibrations in ring structure. These intensities were weak in the coal seams of lower horizon of Barakar Formation (B-I, II, III, V) and gradually increased towards the seams of upper horizon (R-VII to R-X).

The presence of more hydroxyl groups and CH aromatic groups in the seams of upper horizon (R-VII to R-X) could be the cause for high susceptibility of these seams to spontaneous combustion (Text-fig. 13). The seams of Raniganj Formation have shown endothermic peaks in the temperature range 120°-175°C. The bulging nature of the endothermic peaks was quite apparent up to the seam R-III of Raniganj Formation and later sharp peaks were recorded up to the seam R-X (Text-fig. 8) which is in conformity with DTA analysis suggesting similar break down pattern corresponding to temperature at 120°-170°C.



Text-figure 15—Relationship of peroxy complex (o) and peroxy index (+) vs resin distribution on mmf basis in Raniganj Coalfield.



Text-figure 16—Relationship of crossing point temperature vs fluorescing vitrinite and liptinite group of maceral on mmf basis in Raniganj Coalfield.

These structural variations in the chemical bondages are probably related to petrographic composition particularly reactive resin/hydrocarbon material (Text-fig. 13; Table 2).

DISCUSSION

The results have shown a close correspondence between DTA and DTG analysis curves with crossing point analysis of the samples (Text-fig. 7; Table 2). It is further substantiated by reactive resin/hydrocarbon relationship with the crossing point temperature values (Text-figs. 14, 15), resin content versus peroxy complex and modified peroxy index. Similar trends have also been observed in the relationship of crossing point temperature and peroxy complex with fluorescing vitrinite and total liptinite constituents (Text-figs 16, 17). The →

PLATE 6

(All the photomicrographs are in oil immersion under blue light excitation)

1. Sporinite (S) and liptodetrinite (L) of Raniganj coals.
2. Inertodetrinite (I) filled with green reactive resin (GR) hydrocarbon.
3. Green reactive resin/hydrocarbon (GR) oozing out from vegetal matrix.
4. Yellow reactive resin (YR)/hydrocarbon oozing out from vitrite band (V).
5. Yellow reactive resin (YR)/hydrocarbon penetrating along the vitrite cracks. The orange colour thread-like structures are microspores (S).
6. Green colour reactive resin (GR)/hydrocarbon filled in the empty fusinized cells. Fusinized (F) part is black in colour.
7. Yellow colour reactive resin (YR) associated with perhydrous vitrite (V) and fusinized tissue.
8. Yellow to orange colour reactive resin filled along vitrite cracks.
9. Lath shaped greenish resin (R) in vitrite matrix associated sporinite (S)
10. Yellow colour reactive resin (YR)/hydrocarbon filled in sporinite and in vitrite (V) cracks.

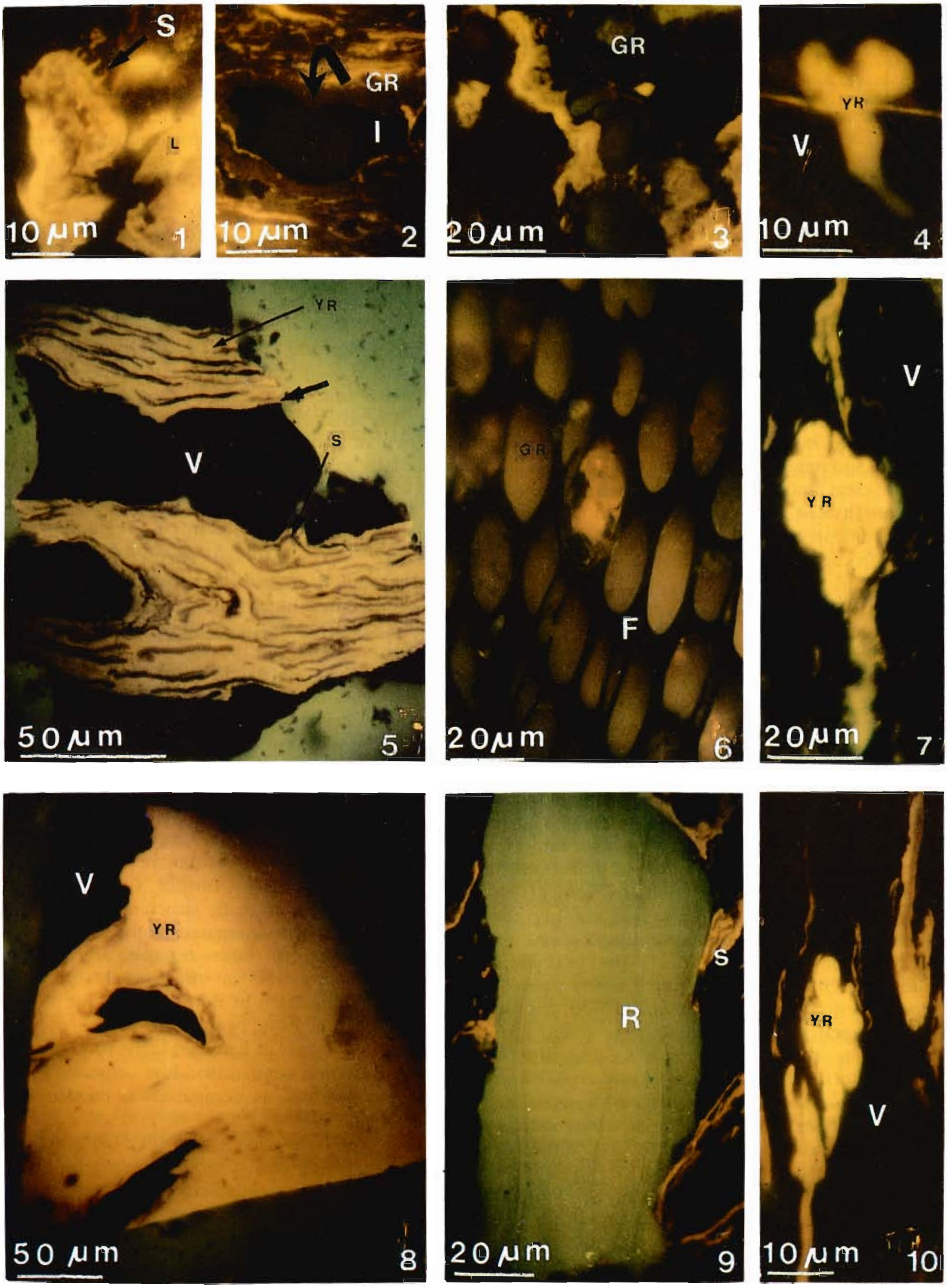
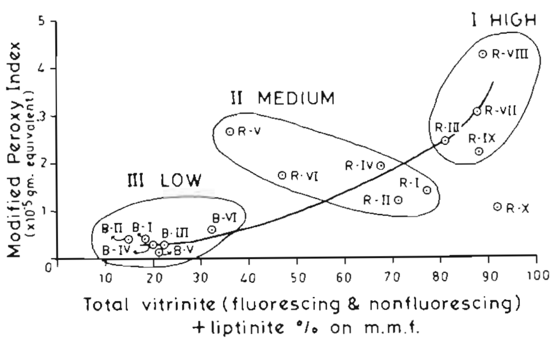

S
L
10 µm
1
GR
I
10 µm
2
GR
20 µm
3
YR
V
10 µm
4
YR
V
S
50 µm
5
GR
F
20 µm
6
V
YR
20 µm
7
V
YR
50 µm
8
S
R
20 µm
9
YR
V
10 µm
10

PLATE 6



Text-figure 17—Relationship of modified peroxy index vs total fluorescing and non-fluorescing vitrinite + liptinite on mmf basis in Raniganj Coalfield.

abundance of reactive resin contents in the source material as well as the expulsion of hydrocarbon at particular rank stage, its spectral properties (Saxena & Navale, MS) indicative of diverse types are highly prone to spontaneous combustion.

The cleat and cracks formed prominently on vitrain layers are filled with reactive resins/hydrocarbons. Recognition of such reactive resin/hydrocarbon material under UV light in small crevices and cracks of vitrinitic layers in coal suggests high mobility of resin/hydrocarbon material susceptible for combustion. The oxidizable and sorptive nature of semibright and bright lithologies is well known (Ferrari, 1938). Also, the organizational pattern of microconstituents of coals helps in easy access of air to reach the internal surface of the coal seams and the mobile resin/hydrocarbon reacts with the oxygen of incoming air and forms peroxy complex which further decomposes and helps in the enhancement of temperature which at reaching ignition point causes fire, leading to spontaneous combustion and burns first, producing charred oxidized resins (Pl 5, fig. 8-Ch). This process continues and becomes uncontrollable due to exothermic reaction and also due to pyrohic nature of coal The perhydrous vitrinite and terpene rich reactive resins prone to oxygen reaction due to their structural configuration leading to self burning of seam were found to be common in the Raniganj seams. The thermal behaviour of coal seams in response to DTA and DTG values, the nature of exothermic and endothermic peaks and the break down steps of these coal seams support the interpretation made on petrographic composition (Table 2).

Contrary to the coal seams of Raniganj Formation, the Barakar coal seams are least susceptible because of the nature of petrographic composition. The dull lithologies, deficiency of bright bands, reactive resin hydrocarbons, cracks and crevices, large amount of inertinitic material together with their high rank nature are some of the properties which decrease the intense reactivity with oxygen resulting in high crossing point temperature and low peroxy complex, hence inhibit the spontaneous combustion. Conspicuously, the resins are totally inert and fusinized (Pl. 5, figs 6, 7, 8) in these coals.

CONCLUSIONS

The coal seams investigated here, vary significantly in their petrographic composition This is in response to coal facies probably controlled by evolving floras and climate. It has been interpreted that the coal seams of Raniganj Formation formed in a particular suite of material and depositional conditions. The characteristic petrographic composition, presence of variety of reactive resins/hydrocarbon particularly green and yellow fluorescing types and their widely dispersed nature, in cleat, small crevices and cracks of bright layer components and along with the empty cells of fusinite as well as in interstitial spaces of coal microconstituents, are probaly strongly amenable to spontaneous burning. Also the organizational pattern of macerals allowing reaccess of air to the internal surfaces due to low rank nature, enhances the spontaneous burning of the coal seams. The Barakar coal seams, developed in relatively toxic conditions as emanated from petrographic composition and dominated by inertinitic coal constituents with more stable and compact lithologies, are least susceptible to spontaneous combustion.

Hence, it is reasonable to presume that the low rank perhydrous vitrinite has imparted the Raniganj coals to be rich in hydrocarbon contents along with reactive resins. This is further substantiated by hydroaromatic, CH aromatic structural configuration of Raniganj coals as compared to the Barakar coal. The availability of these reactive sites in the resin/hydrocarbon or in the mobile phase of vitrinite structure rich in fluorophores due to their low rank nature have been attributed to the high reactivity of oxygen in Raniganj coals forming more peroxy complex and also low crossing point temperature values as compared to Barakar coals (Table 2).

The study postulates that such an integrated analytical system may provide an insight for the better and fuller understanding of the highly debated phenomenon of spontaneous combustion responsible for the mine fire occuring in other Indian coalfields, viz., Jhingurdah, Singrauli, Hutar, Talchir, North and South Karanpura. The study will further help the mining Industry to reorient the mine plan strategies to save the fossil fuel deposits of the country which is being engulfed by the fire due to spontaneous heating phenomenon.

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