Spectrofluorimetric study of some resinites from Indian coals and lignites

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Spectral fluorescence measurements were carried out on three common resinite types—with greenish-yellow, yellow, and orange fluorescence colours, from the Tertiary (Eocene) lignites of Matanomadh and Panandhro Lignite fields of Kutch, and Permian (Lower Gondwana) coals from Raniganj Coalfield. The analysis revealed a distinct shift in their maximum fluorescence intensity (λ max.) towards higher wavelength and increase in red/green (650/500 nm) quotient (Q) corresponding with increase in the age of the deposit. The resinite types from Tertiary lignites show λ max. at 500-510 nm for type-1, 520-530 nm for type-2 and 540 nm for type-3 with corresponding Q values of 0.17-0.29, 0.38-0.49 and 0.69, respectively. Whereas, the Permian coals have λ max. at 580, 600 and 610 nm and Q values of 0.59, 1.47 and 1.9, respectively for greenish-yellow, yellow, and orange fluorescing resinite types.

Key-words-Spectrofluorimetry, Resinites, Lignites, Permian, Eocene (India).

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साराँश

भारतीय कोयलों एवं लग्डांगारों से प्राप्त कुछ रेज़िनाइटों का वर्णप्रतिदीप्ति अध्ययन

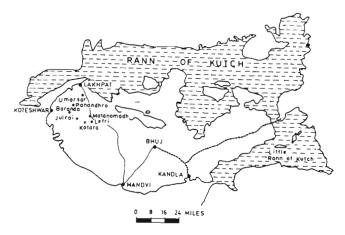
बसन्त कमार मिश्रा, राकेश सक्सेना एवं आनन्द प्रकाश

कच्छ के मातानोमढ़ एवं पनान्धो लगुडांगार क्षेत्रों के तृतीयक युगीन (आदिनूतन) लगुडांगारों तथा रानीगंज कोयला-क्षेत्र से परमी (अधरि गोंडवाना) युगीन कोयले के तीन सामान्य रेज़िनों का वर्णप्रतिदीप्ति अध्ययन किया गया। उक्त विश्लेषण से उच्चतर तरंगदैर्ध्य की ओर अधिकतम प्रतिदीप्ति तथा तिक्षेपों की आयु बढ़ने के साथ लाल/हरे (650/ 500 ऍन-ऍम०) विभाग में भी बृद्धि व्यक्त होती है। तृतीयक लगुडांगारों के रेज़िनाइटों की अपेक्षाकृत परमी यगीन कोयलों में लेमडा अधिकतम का मान तथा विभाग का मान दोनों ही अधिक प्रेक्षित किये गये हैं।

FLUORESCENCE microscopy, a relatively recent innovation for organic petrography, employed in the study of peat, lignite, bituminous coal, oil shale and dispersed organic matter, has been found quite successful and acceptable, particularly in. identification, characterization and quantification of liptinite macerals. It is better suited for rank or maturity assessment than reflectivity measurements in cases where huminite/vitrinite maceral is not present in sufficient quantity or it is unsuited for reflectivity measurements, e.g., sapropelic coals, oil shales and dispersed organic matter.

Quantitative monochromatic fluorescence measurement on coal macerals was carried out by Jacob (1964, 1974). Polychromatic UV fluorescence spectral measurement was initiated by van Gijzel

(1967, 1967a, 1975) and Ottenjann et al. (1975). The technique was later refined and perfected by Ottenjann (1980, 1982). Fluorescence characteristics, being an overall reflection of chemical composition of an organic matter, are related with its rank (maturity) and age (Teichmüller, 1982; Ottenjann, 1982; Teichmüller & Durand, 1983). Macerals, sporinite, alginite, cutinite and suberinite have been studied most, whereas, resinite has not been studied in detail to that extent because of their varying fluorescence properties in a single coal or lignite sample. Characterization and classification of resinites maceral have been carried out by Crelling et al. (1982), Crelling and Bensley (1983), Dobell et al. (1984) and Teerman et al. (1987). Mukhopadhyay and Gormly (1984) found



Text-figure 1-Location map of Matanomadh and Panandhro Lignite field, Kutch, Gujarat.

variation in hydrogen content of resinites with variation in fluorescence colour and property. Recently, Misra (1989, in press) categorized resinites of Panandhro lignite into five types based on their fluorescence spectral characteristics. Therefore, we purposefully selected the resinite maceral from resinite-rich Tertiary (Eocene) lignite and Lower Gondwana (Permian) coal to ascertain possible relationship between the resinite types and their fluorescence properties and age.

LIGNITE-BEARING AREA : KUTCH BASIN

The lignite deposit in Kutch Basin of Gujarat State occurs at several places, viz., Umarsar, Panandhro, Akri-Mota, Matanomadh, Lefri, etc. associated with Palaeocene-Eocene sedimentary sequence lying over the basement of Deccan basalt of Late Cretaceous-Palaeocene (Text-fig. 1). The sequence containing lignite is represented by the sediments of Matanomadh (Palaeocene) and Naredi (Eocene) formations. The Matanomadh Formation is characterized dominantly by gritty, coarse-grained sandstones and shales with occasional, uneconomic, impure lignite occurring as thin streaky or lensoid bands. Whereas the Naredi Formation overlying the Matanomadh Formation comprises shales, carbonaceous shales, sandstones and lignite. The formation houses most of the commercially exploitable lignite present in Kutch Basin, particularly the deposit of the Panandhro Lignite field. The lignite of the Matanomadh area is distributed in eight to nine impersistent to persistent seams, varying in thickness from 0.15 to 4.00 m. In the area of Panandhro Lignite field, the lignite deposit is the largest in the basin. It is represented by three to five persistent seams varying in thickness between 0.10 to 10.51 m being

associated with shale, carbonaceous shale and clay beds. The seams show splitting and merging tendency and become contaminated with other sediments towards south-west (for detailed geology and lithostratigraphy please refer to Biswas & Raju, 1973).

The lignite samples collected from both Matanomadh and Panandhro are from the Naredi sediments. However, the local names have been used in the text mainly because of their occurrence close to the famous Matanomadh temple and the Panandhro Village of the area, respectively.

COAL-BEARING AREA : RANIGANJ COALFIELD, DAMODAR GRABEN

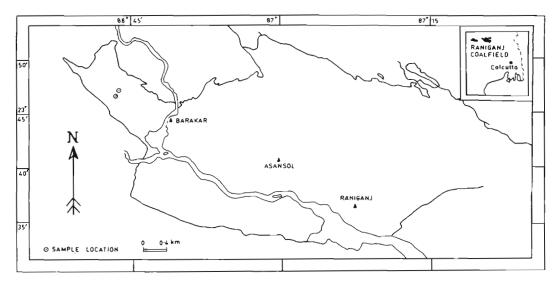
The Lower Gondwana (Permian) sediments of Barakar and Raniganj formations from Raniganj Coalfield of Damodar Graben belong to Damuda Group (Text-fig. 2). The sediments of both the formations are very well developed in this coalfield which represent their type area. The Barakar Formation essentially comprises medium-grained whitish-grey sandstones, shales, carbonaceous shales and low rank non-coking coals, whereas, the overlying Raniganj Formation is characterized by fine-grained sandstones, shales, carbonaceous shales and low to medium rank coking coals. The Raniganj sediments are relatively more argillaceous in nature than that of the Barakar Formation. The coal seams in both the formations generally vary in thickness between 1.50 to 6.00 metres.

For the present study the Barakar coal from Gopinathpur, Chanch-Begunia and Brindabanpur seams and Raniganj coal from Koithi and Handal seams have been selected as they contain fair amount of resinite maceral for spectral fluorescence measurement.

MATERIAL AND METHOD

Eighteen representative channel samples from Matanomadh (3 samples) and Panandhro (10 samples) lignite and Barakar (3 samples) and Raniganj (2 samples) coals have been used for the present investigation. Particulate pellets from crushed and sieved (\pm 18 mesh, 1.00-2.00 mm grain size) lignite and coal samples were prepared by cold embedding in Epoxy resin. The particulate pellets were ground and polished following international standard methods. The descriptive terminology and quantitative assessment of coal and lignite macerals (microconstituents) are according to I.C.C.P. recommendations (I.C.C.P., 1971, 1975).

Spectral fluorescence measurements on resinite



Text-figure 2-Location map of Raniganj Coalfield, West Bengal.

were carried out between 400 to 750 nm, with 10 nm interval using 150 watt xenon lamp as the illumination source on Leitz MPV-3 unit. NPL Fluotar oil objective of 25 magnification (0.75 numerical aperture) was used with fluorescence-free immersion oil. A Littrow type grating monochromator (12001/mm) on emission side $(\lambda 2)$, central control panel and a bench-top Hewlett-Packard 85B computer was attached to the MPV-3 unit. Monochromator drive, shutters and measurements were controlled by central control panel or the computer (one at a time). Scanning of samples was done in blue light (420-490 nm, violet blue excitation), whereas UV excitation filter was used (340-380 nm) for quantitative spectral measurement. Correction factor from calibration spectrum was obtained using Planck's colour temperature distribution (at 3000° K) of stabilized 12 volt 100 watts tungsten halogen lamp in transmitted mode (Reuter et al., 1976; Ottenjann, 1980). All measurements (background & calibration of sample spectra) and calculations (background substraction, correction factor, spectral corrections, λ max, and red/green quotient) were performed with the help of the attached computer including

print-out of the result and curve. For operating the MPV-3 and computer, operating manual and programmed cassette for spectral measurement were supplied by Leitz. All necessary precautions were taken during measurements, e.g., selection of site for background measurement and object for spectral measurement avoiding alteration and weathering effects. Erroneous results due to any possible source were rejected.

A minimum of ten measurements were carried out on each resinite type. Individual fluorescence spectral data were averaged and normalized to 100 per cent (λ max.) and average curve for resinite was drawn manually.

MEGA- AND MICROSCOPIC CHARACTERS OF LIGNITES AND COALS

Lignite—The lignite from both the Matanomadh and Panandhro areas of Kutch are dark-brown in colour, sparingly banded in nature and amorphous in texture. However, Matanomadh lignite is much inferior in quality as compared to the Panandhro lignite. They contain frequent globular or lensoid bodies and bands of yellow and red-coloured resins,

PLATE 1

(All photomicrographs are under blue light excitation)

- 4. Cutinite with yellowish-orange fluorescence colour in Panandhro lignite, × 672.
- 1. Resinite type-1 (greenish-yellow) in Panandhro lignite 5 (R-other resinite types, Sp.-pollen grain), × 672. 6
- 2. Resinite type-2 (bright yellow) in Matanomadh lignite (R-other resinite types), × 672.
- 3. Resinite type-3 (orange) in Panandhro lignite, × 576.
- Resinite type 1 (greenish yellow) in Permian coals, × 576.
 Resinite type 2 (bright yellow) in Permian coals (Sp.—Spori-
- nite), ×416.
- 7. Bright yellow fluorescing sporinite (Sp.) in Permian coals, × 672.

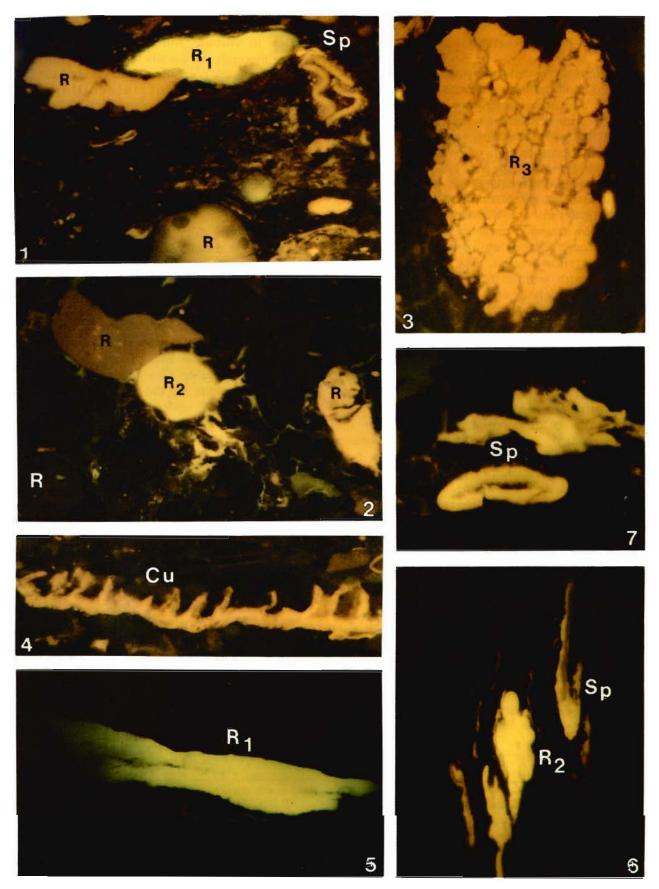


PLATE 1

dispersed throughout the seams. Biogenic (framboidal) pyrite is commonly associated with the lignite.

The lignites, in reflected white light, are rich in huminite and liptinite macerals. The inertinite macerals, consisting chiefly of semifusinite and sclerotinite, are less in amount. The mineral matter content is generally low to moderate, but occasionally becomes high, when lignite turns shalv in nature. Framboidal pyrite and calcite (both concretionary and secondary) are common, whereas argillaceous black granular mineral matter is usually in subordinate amount. Under blue light both the lignites show common presence of maceral alginite (Botryococcus) fluorescing with bright, greenishyellow to yellow colour. The sporinite maceral fluoresces with whitish to yellow colour and is distributed sparsely as well as in occasional dense patches (Pl. 1, figs 1, 6, 7). Cutinite is common to quite common and fluoresces with orangish brown, orange, yellowish brown, or brown colour (Pl. 1, fig. 4). Suberinite maceral is sporadic to common fluorescing with weak yellowish or orangish-brown to brown colour. The macerals fluorinite and exudatinite are only sporadic. The maceral liptodetrinite is quite common in most of the samples studied. Among the macerals of liptinite group, resinite is the abundant maceral in both Matanomadh and Panandhro lignites.

The resinite in these lignites, occurs as primary globular, oval or elliptical discrete bodies, primary cell filling and as remobilized lumpy or irregular elongated bodies. It shows brown, shades of grey and dark reddish-brown colour under normal reflected light. Under blue light excitation some of the resinites display droplet or vesicle structures and cracks. The resinites show yellowish-green, greenish-yellow, orangish-yellow, yellowish-orange, orange, brownish-orange, orangish brown, yellowish-brown to brown fluorescence colours. However, for the pesent study only yellowish-green to greenish-yellow (resinite type-1), yellow (resinite type-2) and yellowish-orange to orange (resinite type-3) fluorescing resinites as classified by Misra (1989, in press) from Panandhro lignites have been selected.

Coal—The coals of Barakar and Raniganj formations from Raniganj Coalfield are banded in nature. The Barakar coals have frequent and prominent dull bands alternating with impersistent semi-bright and bright bands. The Raniganj coals, on the contrary, have persistent bright and semi-bright bands with thin, dull bands. Both the coals are hard and compact in nature and break with uneven fracture

Under reflected white light, the Barakar coals are rich in vitrinite and inertinite macerals. The liptinite macerals are usually less in proportion. The Raniganj coals are normally rich in vitrinite macerals, chiefly consisting of telocollinite and desmocollinite. The liptinite macerals, particularly resinites, are more common than the Barakar coals. The inertinite macerals mainly comprising semifusinite, fusinite and inertodetrinite are seldom represented by more than 25.00 per cent by volume. In both the Barakar and Raniganj coals, cell lumens of semifusinite and fusinite are often filled with argillaceous or calcareous minerals. In general, these coals have high amount of mineral matter consisting mainly of black granular matter (argillaceous mineral matter). Calcite and siderite minerals are common and may become occasionally high. The Raniganj coals, in this area, are characterized by finely disseminated and crackfilling pyrite occurrences.

Under blue light excitation, the liptinitic contents in both the Barakar and Raniganj coals increase considerably (2-4 times). The sporinite, resinite and cutinite are the most common liptinite macerals observed under fluorescence light. The exsudatinite is common and suberinite is usually sporadic in Barakar coals. However, the coals from Raniganj Coalfield are characterized by the presence of liptodetrinite and relatively high proportion of resinite macerals. The sporinite and cutinite show vellowish-orange and orange fluorescence colours. The resinite fluorescing with green, greenish-yellow, bright yellow, yellowish orange, orange, orangishbrown and brown colours occur as primary globular, oval or elliptical discrete bodies as well as remobilized crack and fissure fillings.

RESULTS

The averge spectral parameters of resinite type-1 (yellowish-green to greenish-yellow), type-2 (yellow) and type 3 (yellowish-orange to orange) recorded in Tertiary lignites and Permian coals are listed as follows (Text-figs 3.8):

λ Max.	Red/Green Quotient (Q)
500	0.17
530	0.379
510	0.29
520	0.49
540	0.69
	500 530 510 520

Permian resinite

Perman resince		
LR-1	570	0.588
LR-2	590	1.29
LR-3	610	1.90

The description of qualitative and spectral fluorescence characteristics of resinite types is as follows:

Resinite Type-1 (Pl. 1, figs 1, 5)—The resinite type-1 (yellowish-green to greenish-yellow) occurs as oval, elliptical or globular discrete bodies distributed sparsely with other resinite types. This resinite is less common in Tertiary lignites and sporadic in Lower Gondwana coals. In lignites these resinites are translucent in appearance under blue

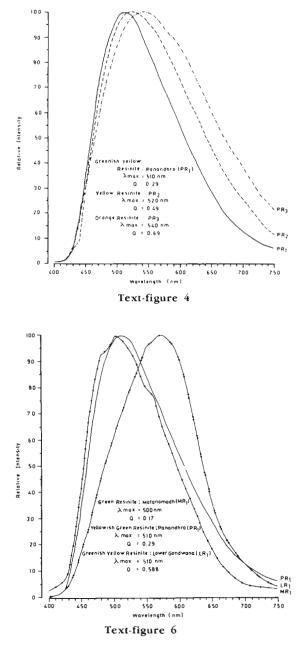
90 80 70 60 Intensity 50 2.010114 40 Residule - Matao λ max = 500 nm Q = 0 17 30 esin:te μR₂ λ mox = 530 nm 20 Q = 0.379 10 N P ٥ 450 500 5 50 600 650 700 750 (nm) Text-figure 3 100

90 80 70 Relative Intensity 60 50 40 sinit • 30 λmax + 570 nm 0 = 0.588 esinite : LR2 λmax = 590 nm 20 Q = 1.29 esinite : LRa 10 ۵ 1.9 0 -200 450 500 550 600 750 650 700 Wavelength (nm) Text-figure 5

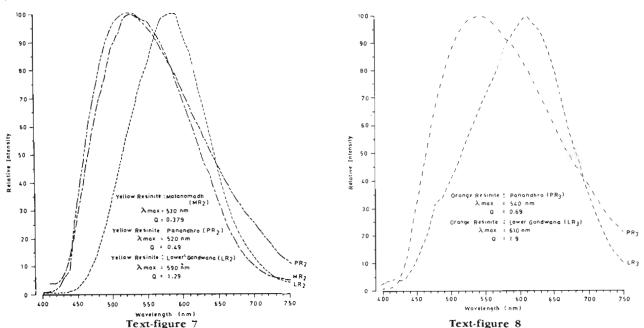
light excitation. They do not show droplet or vesicled structures; however, occasionally they are cracked.

The average λ max. of resinite type-1 in lignites from Matanomadh and Panandhro has been recorded at 500 and 510 nm with Q values of 0.17 and 0.29, respectively. For Permian coals the λ max. has been found at 570 nm with red/green quotient (Q) of 0.558.

Resinite Type-2 (Pl. 1, figs 2, 6)—The resinite type-2 (yellow) is common in Tertiary lignites and Permian coals. It commonly occurs as discrete bodies (like type-1) and also as infillings in cell lumens of humotelinite in lignites. In Lower Gondwana coals cell filling resinite type-2 has not



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Text-figures 3 to 8—Spectral fluorescence pattern of: 3, Green (MR1) and yellow (MR2) resinite types from Matanomadh Lignite, Kutch; 4, Greenish yellow (PR1), yellow (PR2) and orange (PR3) resinite types from Panandhro lignite, Kutch; 5, Greenish yellow (LR1), yellow (LR2) and orange (LR3) resinite types from Lower Gondwana coals, Raniganj Coalfield, West Bengal; 6, Resinite type-1 from Matanomadh and Panandhro lignites and Permian coals; 7, Resinite type-2 from Matanomadh and Panandhro lignites and Permian coals; 8, Resinite type-3 from Panandhro lignite and Permian coals.

been recorded. In lignites, this resinite type appears translucent. In coals, its fluorescence intensity is weaker than those of the lignites. Cracks and droplet structures on these resinites are uncommon.

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The average maximum intensity (λ max.) of the resinite type-2 occurs at 530 nm for Matanomadh, 520 nm for Panandhro and 590 nm for Lower Gondwana coals with red/green quotient of 0.379, 0.49 and 1.29, respectively.

Resinite Type-3 (Pl. 1, fig. 3)—The resinite type-3 (yellowish-orange to orange) is common in Tertiary lignites and quite common in Permian coals. It occurs as discrete bodies, like type 1 and 2, and also as irregular lumps. In lignites it is rarely found as cell fillings. In lignites, this resinite type under blue light excitation commonly shows cracks and droplet structure. In Permian coals droplet structure on resinites has not been observed. These resinites are occasionally transluscent in lignites. The fluorescence intensity of these resinites is weaker than type 1 and 2.

The average maximum intensity (λ max.) of the resinite type-3 occurs at 540 nm in Panandhro lignite and at 610 nm in Permian coals. The corresponding red/green quotients are 0.69 and 1.90, respectively.

DISCUSSION

The three resinite types in Tertiary lignites of Matanomadh and Panandhro, and Permian coals of

Raniganj Coalfield, Damodar Graben are distinguishable using two spectral parameters— λ max. and red/green (650/500 nm) quotient (Table 1). The three resinite types (only two from Matanomadh lignite) of lignites and coals separately show only small differences in their spectral parameters (Text-figs 3-5). They may represent a continuous series as expressed by Teerman *et al.* (1987) and Misra (1989, in press).

Certain physical characters, viz., droplet (uncollapsed gas bubbles) or vesicle (collapsed bubbles) structures and transluscency in resinites, have not been observed in the Permian coals. Absence of these features in Permian coals and their presence in lignites is possibly related to the vegetal precursors which form lignite and coal. The resinites in Tertiary lignites originated chiefly from angiospermic vegetation and those in Permian coals were produced from gymnospermic plants (Stach *et al.*, 1982). The absence of transluscency in resinites of Permian coals may also be related with their older age and higher rank.

Comparison of spectral curves of the individual resinite type with each other (Text-figs 6-8) distinctly shows a shift in maximum fluorescence intensity (λ max) towards higher wavelength with corresponding increase in red/green quotients. The resinite types from lignites, being of identical age (Eocene), appear to be closely related with each other (Text-figs 6-8) by virtue of similarities in their spectral pattern, λ max. and red/green quotients. However, certain minor differences in their spectral characteristics may have been caused by the differences in rank, conditions of coalification and tectonic history of the two lignite deposits. Thus, the evident shift of λ max. towards higher wavelength along with increase in red/green quotient in each of the resinite types from Tertiary to Permian times corresponds fairly well with age of the lignite and coal deposits.

CONCLUSIONS

The spectral fluorescence characteristics of the three resinite types, type-1 (yellowish-green to greenish-yellow), type-2 (yellow), and type-3 (yellowish-orange to orange) recorded in Tertiary lignites from Matanomadh and Panandhro areas of Kutch and Lower Gondwana (Permian) coals from Raniganj Coalfield of Damodar Graben suggest that:

(i) the three resinite types from different areas show gradational spectral characteristics among themselves and may represent a continuous series;

(ii) the Palaeocene-Eocene resinite types in Matanomadh and Panandhro lignites being identical in age are also closely related on the basis of their spectral pattern and other parameters;

(iii) the three resinite types from Tertiary lignites and Permian coals show a distinct shift in their maximum fluorescence intensity (λ max.) towards higher wavelength and corresponding increase in red/green quotients. The distinctly higher λ max. and higher red/green quotients of the three resinite types of Permian coals vis-a-vis those of the resinite types of Tertiary lignites appear to be related with the age of the deposits.

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