Greenfield Energies from Underground Coal Gasification and Liquefaction of Solid Fossil Fuels—Basics and Future Potentiality in India

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1 Introduction

Coal has a complex macro-molecular network, consisting of polyaromatic clusters linked with side chains and functional groups on peripheral positions by covalent and non-covalent bonds [1]. The monomer organic compounds trapped in the porous structure of coals may be products and residues of the coalification process, which are thought to be related to the macromolecular precursor material. A spate of models has been proposed for macromolecular structure of coal based on experimental studies on structural and chemical analysis of coal [2]. Chemical identification of functional groups and general information on the carbon skeleton are established, but the structural examples given must be seen as attempts only to give the general framework of the coal structure. They are mainly intended to describe structures of hard brown coal to bituminous coal. The aromatic centres are generally linked by hydroaromatic and methylene bridges and fringed with methyl, hydroxyl, carboxyl, keto, amine and other functional groups.

Petrographic characteristics of coal play a key role in defining its suitability for its various industrial applications. Thus, importance of coal characterization is well established and is crucial for making metallurgical-quality coke for blast furnace applications, in thermal power plant electric power generation, coal beneficiation, hydrocarbon exploration and assessments including coal bed methane, underground coal gasification and coal liquefaction. Maceral composition as well as rank of a coal has got a tremendous significance in determining the quality of coal and suitability for specific applications. Coal is composed of microscopically distinct components of the organic matter, called macerals, derived from terrestrial, lacustrine and marine plant remains. They differ in their physico-chemical and

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U. K. Bhui (ed.), *Macromolecular Characterization of Hydrocarbons for Sustainable Future*, Green Energy and Technology,
https://doi.org/10.1007/978-981-33-6133-1_13

optical properties such as hydrogen content, aromaticity, reflectance, density. These are grouped as: (i) vitrinite, with intermediate reflectance and high oxygen-to-carbon ratios, derived from woody tissues; (ii) liptinite, with low reflectance and high hydrogen-to-carbon ratios, derived from spores, cuticles, resins and algal bodies and (iii) inertinite, with high reflectance and carbon contents, derived from fossil charcoal or decayed material. Hydrocarbon generation potential of coals are dependent on the amount, type and maturity of the inherent organic matter present within them [3, 4]. This organic matter within the coal changes from one lithotype to another with progressive changes of macerals composition [5–8]. Individual macerals and their associations in lithotypes may play different roles in generation and storage potentiality. Therefore, understanding the maceral compositions of lithotypes is important for gaining insights into their behaviour during hydrocarbon generation and storage.

In the present work, an attempt is made to understand the basic factors responsible for a favourable utilization of solid fossil fuel towards conversion to gas or oil. Based on the analysis of the available subsurface and surface data, either accrued through exploratory drilling by GSI or generated through researches all over the country, site-specific possibility has been explored for use of coal or lignite for UCG and CTL. The study for UCG was done with the physical, chemical, structural and petrographical data of the coal/lignite, while for CTL, petrography, thermogravimetric analysis and differential thermal analysis (TGA-DTA), Fourier transform infrared (FTIR) spectroscopy, UV–visible and fluorescence spectroscopy have been carried out to obtain the precise knowledge of macromolecular structure and composition for better site-specific and technology-specific utilization towards green and clean energy.

2 Underground Coal Gasification (UCG)

Underground coal gasification (UCG) is a technique of converting unrecoverable and unmineable coal into a combustible gas, which can be used for industrial heating, power generation or the manufacture of hydrogen, synthetic natural gas or diesel fuel. It allows to fully utilize the coal resource from otherwise unrecoverable coal deposits in an economically viable and environmentally safe way. UCG turns this resource into high-value products like clean power, liquid fuels, syngas, fertilisers and other chemical [3–6]. UCG has the potential to unlock vast coal energy resources that cannot be beneficiated economically through conventional mining methods. In the process coal, oxygen and water/steam are reacted at high temperatures through chemical reactions in the underground gasifier in situ to form synthesis gas (syngas), predominately being CO, H_2, CH_4 and CO_2. A UCG operation consists of a series of injection and production wells drilled into a coal seam where reactant gases, i.e. air, oxygen or steam, are injected to cause the gasification by partial combustion instead of complete combustion of coal (Fig. 1). Besides primary gas components, a number of organic and inorganic contaminants, such as phenols, benzene, sulphates, metal and metalloid elements, can be generated and released during the UCG process [9]. Simplicity of UCG is very attractive, but development of reliable working programme with acceptable cost benefit ratio seems difficult in practice. Technical feasibility depends on multidisciplinary aspects; thus, trials must be undertaken at pilot scale. For every geological set up, a tailor-made site-specific work plan has to be designed to exploit the targeted resource. UCG is an inherently clean coal technology as it reduces sulphur and nitrogen oxide emissions to very low levels and low volatile organic components, methane and greenhouse gases (GHG) emission to atmosphere. Total solid waste from UCG is typically half the volume generated by conventional coal plants and water use is substantially lower as well. UCG technology was first developed in the Former Soviet Union (FSU) in the 1920s. The USA and Europe also conducted several tests in the 1970s and 1980s. In the 1990s, China began UCG research and development and is continuing with this effort.

The first consideration is the selection of the site, which must take into account the condition and quality of the coal, the permeability of surrounding strata, the proximity to aquifers and the geological and hydrogeological conditions. This is a complex area, requiring detailed knowledge of site geology by exploratory drilling, seismic surveys, laboratory testing and modelling.

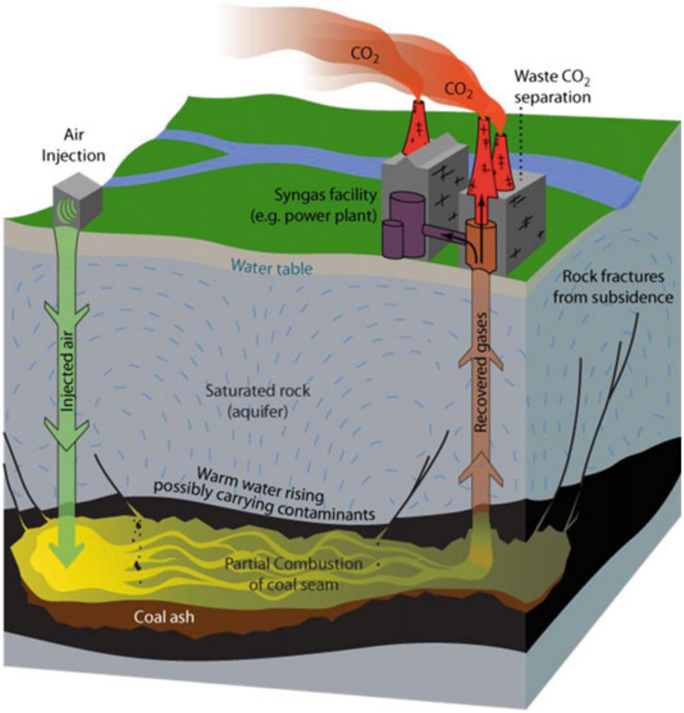


Fig. 1 Void formation during UCG process (*Source* Water Cartoon, Kissclipart.com)

2.1 Desirable Coal Characters

Coal with higher reactivity having significant amount of exinite/liptinite, high moisture content which facilitates reactions providing water, low-rank coal can be easily gasified. Caking and swelling index should be less; low mineral matter and ash fusion temperature and adequate permeability are important parameters for UCG treatment.

2.1.1 Physical Attributes of Target Coal Seam

(i) Depth of occurrence should be more than 100 m and preferably less than 300 m, (ii) thickness of coal seam should be more than 2.5 m with thickness variation less than 25%, and (iii) optimum permeability of the coal seam is desired as 50–150 md.

2.1.2 Chemical Properties of Target Coal Seam

(i) Ash content of coal seam should vary from 10 to 40% [heat value/calorific value (3.5–4 MJ/m^3] of product gas almost constant up to 40% of moisture content, beyond which steadily declines. (ii) Low-rank coal with relatively high moisture and porosity is preferred, desirable moisture content should be less than 15 wt%. (iii) Desirable sulphur content should be less than 1 wt%. (iv) Coal with low caking and swelling indices is preferable for UCG, and higher values result instability of gasifier. (v) Reactive macerals increase amount of hydrogen content in the product gas. Higher the liptinite content, higher will be the H/C ratio, which produces large amount of gas on heating. Low-rank coal with high inertinite and liptinite macerals are suitable because it suppresses excessive caking and swelling which are not desirable.

2.1.3 Geo-Mechanical Character of Coal and Associated Strata

Coal and strata density, compressive and tensile strength, modules of elasticity, Poisson's ratio, angle of internal friction, cohesion, porosity, permeability ratio, properties of clay and sands, plasticity number and liquid limit for clay and grain size analysis are required for modelling any UGC project, which are available from drill core study.

2.1.4 Structural Set-up

Higher dip of coal seam is preferred; steeper gradient enhances better pyrolysis producing more gas. Tars/liquids flow down in steep dipping beds favouring effective combustion.

Joints and faults are the discontinuities in coal seam which are identified based on surface and subsurface geological data. Coal seam targeted for UGC should be considerably away from identified major faults. Precise characterization of joints and faults as well as the mine-induced fractures within the coal/lignite seams is to be taken care of.

2.1.5 Overburden Characteristics

Impervious strata in roof and floor of seam is preferred, desirable thickness of the litho assemblage overlying targeted coal seam for UCG should not less 15 m which is to be devoid of any workable coal seam. Desirable permeability of overburden should be <5 md, having considerable mechanical strength to avoid roof collapse.

2.1.6 Hydrological Regime

No good aquifer should be in the vicinity of UGC project so that any quality groundwater supplies will not be put at risk. Thorough characterization of existing aquifer of the surrounding area is a must. Careful monitoring during operations, particularly maintenance of optimal pressure always less than hydrostatic pressure in gasifier is extremely necessary. Overlying aquifers must be sufficiently separated by an impermeable seal to prevent either excessive water inflow to the gasifier or gas escape to the overlying aquifer through any fractures that develop above gasifier. Similarly, underlying aquifers must be sufficiently distant to prevent excessive water inflow to the gasifier from deeper and higher pressure zones.

Groundwater pollution is considered to be one of the most serious environmental issues [10–13]. This may be caused by the dispersion and penetration of gaseous products into surrounding rock layers due to the development of higher pressure than hydrostatic pressure in gasification process to prevent the influx of groundwater, the emission and dispersion of contaminants with gas products and migration of gasification residue by leaching with the penetration of groundwater after gasification. The physicochemical properties of groundwater may be affected by the hazardous escaped gases. Phenolic compounds are major organic contaminants in groundwater, PAHs and heterocyclic compounds are minor organic contaminants of concern, and a wide array of ionic species, such as calcium, sulphate, bicarbonate, chloride, sodium and ammonium, etc., are also present in larger amounts [12]. Solid residue remained underground after UCG could be a source of contaminated groundwater [10, 14].

2.1.7 Land Use Considerations

Every geological setting is different, and the surface constraints vary due to land use, surface topography and other factors. The site of UGC project should be at outskirt with limited human activity, settlement or mine. No waterways (rivers/

lakes) should overly the site for UGC project. Field development plans and surface facilities layout should be studied to ensure the lowest lifecycle cost of the produced syngas.

2.2 Basics of UGC Process, Chemical Reactions and Void Formation

Higher coal quality, deeper coal formations targeted for UCG and lower water ingress results in improved oxygen utilization. Oxygen blown gasification is significantly more efficient in utilizing the injected oxygen as compared to air blown gasification and produces a syngas with a high-quality suitable for petrochemical applications. The gasification process requires high temperatures to convert the coal into syngas. In the gasification zone, temperatures exceed 1000 °C and may reach 1500–2000 °C when pure oxygen gasification is applied. The three essential types of char layer reactions which govern the UCG product gas composition are oxidation, reduction and pyrolysis (Fig. 2). In the oxidation zone, wet coal is heated and converted into dry coal by removing moisture attached to it. Upon further heating of the coal, the pyrolysis reactions begin at temperatures around 350–400 °C [11, 15] and coal loses its weight, generating volatile matters and the solid known as char. Finally, the char reacts with the injected/pyrolyzed gases to produce the syngas [15]. Empirical and exact mathematical tools and approaches can be used to provide insights into the underground processes through experimental cavity modelling [16–18]. The outflow channel is divided in small sections along the length, each consisting of rubble zone, void zone and roof at the top, which are well consistent with experimental models.

2.3 Environmental Impact

Environmental pollution in the form of generation and transport of contaminants via experimental and numerical analyses is the dominant issue. Leaching of organic substances from the gasifier, e.g. phenol, increased concentration of inorganic salts near the gasification zone dissolving of hazardous gases (H_2, CH_4, CO_2, H_2S and NH_3) in ground water, and leaching of heavy metals (Hg, As, Pb, Cr, Cd), emission of pollutants and greenhouse gases to the atmosphere are some of the possible contaminant during UCG operation. A spate of research work has been done in this respect, and possible measures have also been suggested [5, 12, 19–22] Several models with various parameters like modelling of contaminant transport and reactions [22, 23], general hydrogeological models were proposed for mitigation of the adverse effects of UGC.

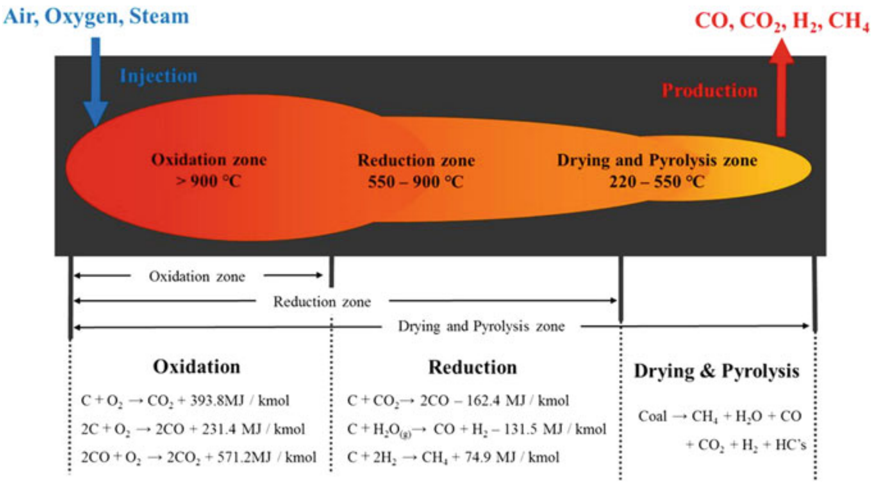


Fig. 2 Typical chemical reaction zone during UCG process [15]

2.4 Surface Subsidence and Effect on the Permeability of Overburden

Removing coal from the underground by mining, UCG can cause overburden to collapse, fracture and strain in the form of surface subsidence, which may likely to change the permeability field. In large-scale UCG operations, it is more likely that higher permeability would be created far above the seam. This can make groundwater contamination worse. To isolate the UCG operation environmentally from shallower sensitive contaminant receptors, the zone of enhanced permeability must stay below the impermeable barriers that are counted on to protect shallower aquifers. Surface subsidence can be mitigated by appropriate site selection, including depth and strength of rock volume; spacing of UCG reactors to leave walls and pillars between produced zones, and identification and avoidance of structural weaknesses as pre-existing faults. Gasification of thick seams may result in a greater vertical extent of roof collapse (goafing). This can open up gas connectivity far up into the fractures of a sagging overburden, making it more likely for product gas to escape up into shallow strata.

2.5 Advantages of UCG

Lower emissions of particulates, reduced methane emissions, SO_2 and NOx, no noise and visual impact on the surface and disposal at mine sites and power and station sites, less transport cost, reduction of operating costs and surface impacts, occupation of smaller land area, lower risk of surface water pollution, non-requirement of mine water recovery, elimination of mine safety issues, and surface hazard liabilities on mine abandonment are some of the advantages of UCG over coal mining. Besides, conventional gasification facilities are not needed in UCG process which involves a significant reduction of capital costs. Most of the ash in coal stays underground during UCG process, thereby avoiding the need for additional syngas clean-up, and the environmental issues associated with ash storage. Commercial UCG applications use substantially less water than conventional gasification technologies.

2.6 UCG, Greenhouse Gases and CO_2 Capture Storage (CCS)

Efforts were made globally effort to understanding UCG's advantages and disadvantages with respect to greenhouse gas (GHG) emissions. Methane comprises a significant fraction of UCG product gas. Methane is a much more potent greenhouse gas than carbon dioxide. Potential leaks of methane containing product gas from UCG operations need to be included in analyses. Gasification is technically amenable to efficient separation and CCS that could then be sequestered (Fig. 3). This has been advanced for surface gasifiers in recent developments and demonstrations. UCG could work similarly, although the methane in UCG's product gas cannot easily be water-gas shifted, limiting the extent of CCS that can be achieved. More researches about carbon capture from UCG syngas and environmental protection are needed with the objective of its application in terms of comparison of global GHG emissions with the fossil fuel and renewable alternatives [20]. There may be sites where the coal is adjacent to a suitable CO_2 store, like a depleted gas reservoir or saline aquifer.

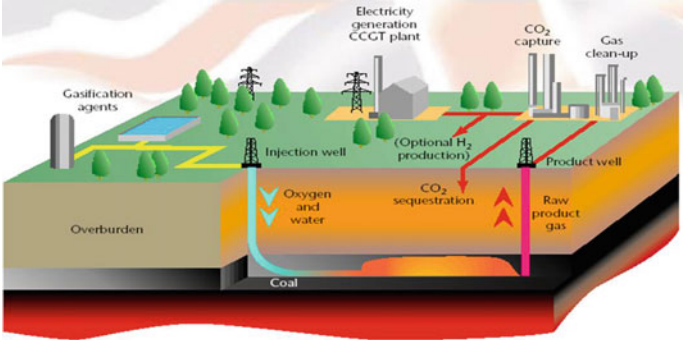


Fig. 3 Site characterization and selection of pilot plant for UCG (*Source* Online published in a blog 'Mining methods in India'; on June 04, 2011

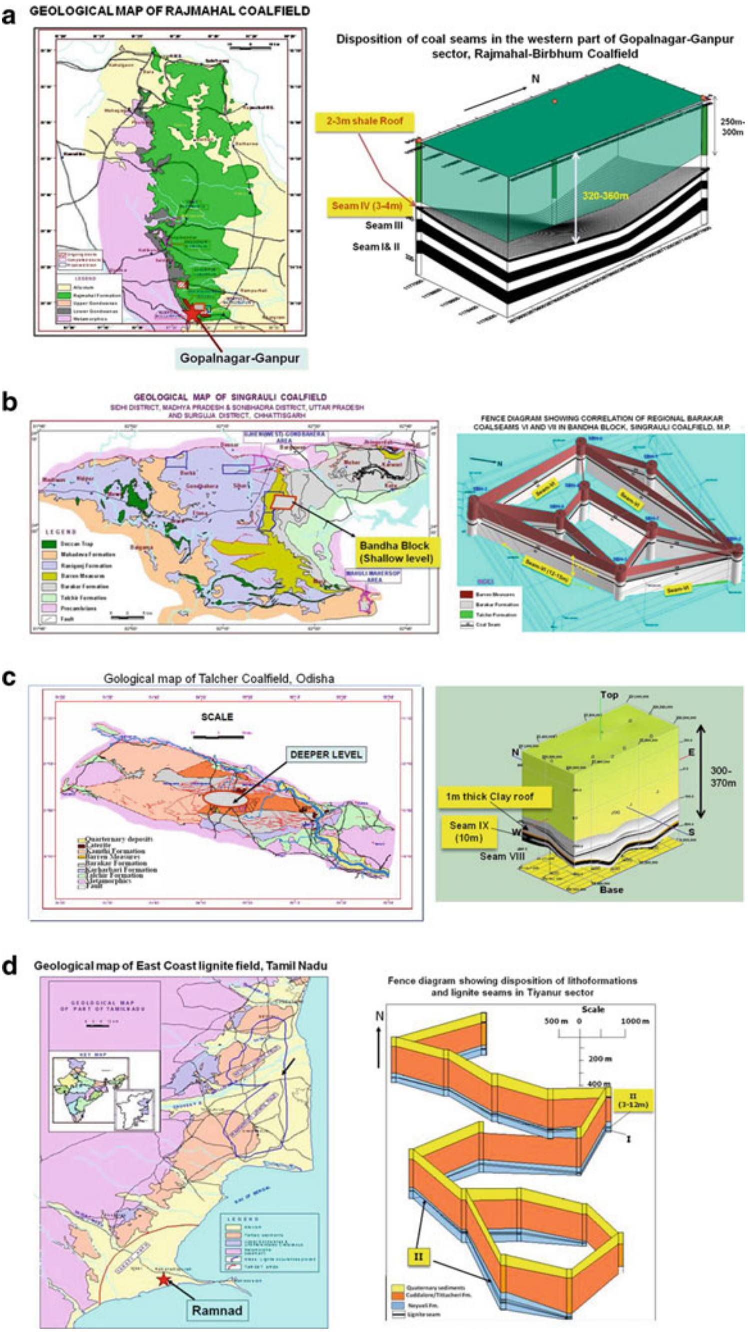


Fig. 4 **a** Gopalnagar–Ganpur block, Rajmahal coalfield and stratigraphic disposition of seam-IV. **b** Bandha block, Singrauli coalfield and fence diagram of seam-VI. **c** Jalatap block, Talcher coalfield and stratigraphic disposition of seam-IX. **d** Tiyanur block, Ramnad Sub-basin and fence diagram of seam II

2.7 *Future Potentiality of UCG in India*

The strategic case for UCG lies with the plentiful supply of indigenous coal, the high cost of fossil fuel alternatives like natural gas and the opportunities it presents for new industry. An instrumented pilot test in the target coal is essential in order to obtain the data to plan, design and operate the commercial application. The choice of technology for UCG is still wide open. Long periods of gasification have been achieved with controlled retracting injection point (CRIP) or the Chinese and Russian methods of multiservice wells. Based on the available surface and subsurface data in and around the targeted coal/lignite seam, choice of technology for UCG should be site specific. Geological Survey of India has created a vast subsurface database of the coal and lignite seams in all the Gondwana and Tertiary coal and lignite fields by exploratory drilling through decades. In-depth analysis of structural, chemical and coal/lignite petrographic data of individual coal/lignite seams has imparted UCG suitability of some of these. Characteristics and essential parameters required for such seams are given here, which warrant specific technologies for taking up UGC projects in future.

Out of the total resource of 32.6 billion tonne, about 13.8 billion tonne of coal (42%) occurs beyond 300 m depth and the total resource of lignite stands as 4.5 billion tonne (as per Inventory of Indian Coal and Lignite Resources as on 01-04-2019), which indicates that there are ample potentiality of UGC projects in the country, considering the site-specific favourable parameters for their implementation.

Based on the exploration activities carried out by GSI in India, some of the characters of the targeted coal/lignite seams are given in Table 1. In addition, some coal seams having the parameters favourable for UCG may also be targeted from Hasdo-Arand, East of Kothagudem of Godavari and Ib-River coalfields, where either embargo on mining exists due to forest cover and are not suitable for immediate mining. Suitable choice of gasifiers and site-specific implementation of pilot projects are mandatory for amplifying this unconventional green energy resource to occupy major space of ‘Energy Basket’ of India keeping pace with the outer world.

3 Coal Liquefaction (Coal to Oil CTL)

Liquefaction of coal to generate synthetic oil is a domain of constant research, and it is generally presumed to proceed via breaking the weak bonds followed by stabilization of the resulting reactive fragments with hydrogen. Depending on the structural bonds present in coal, a sequence of bond breaking process eventually produces reactive molecules while many of the stable bonds remain intact. The liquid product of the liquefaction should contain molecules that are a vestige of structural arrangements present in the original coal. Coals may contain significant quantities of the hydrogen-rich and hydrocarbon-generating maceral group, liptinite. When heated, coals produce petroleum-like pyrolysis products. Bituminous

Table 1 List of some of coal/lignite seams recorded in areas explored by GSI warranting attention for UCG

Coal/ lignite basin	Rajmahal Basin	Singrauli Basin, Son valley	Talcher Basin, Mahanadi valley	Ramnad Sub-basin, East coast lignite
Area	Gopalnagar–Ganpur [24] (Fig. 4a)	Bandha [25] (Fig. 4b)	Jalatap [26] (Fig. 4c)	Tiyanur [27] (Fig. 4d)
Seam no.	IV	VI	IX	II
Thickness (m)	3–4	12–15	10	3–12
Depth (m)	320–360	125–200	300–370	340–460
Roof lithology	Sh: 2–3 m, T. O. > 20 m	Sst: 20 m	Clay: 1 m, T. O. > 15 m	Clay > 20 m
Dip	20–40	20–30	30	20–30
Rank	HVB	HVB	HVB	Lignite
Ash (%)	20–49	23–27	15–45	5–15
Moisture (%)	4–6	6–9	2–13	45
Maceral (%)	L(4–16), I(22–44)	L(16–19), I(32–42)	L(21), I(26)	L(2–10), I(5–10)
Resource (Sq km)	20(4)	235(4)	125(4)	150(28)

Sh Shale, *Sst* Sandstone, *T.O.* Total Overburden, *HVB* High Volatile Bituminous, *L* liptinite, *I* Inertinite

coals contain bitumen that can be easily released by extraction using common solvents. Some coals are oil prone, whereas others are not potential for generating liquid hydrocarbons. Role of microconstituents of coal and corresponding changes affecting its macromolecular arrangement influences the potentiality of generation and expulsion of oil from coal.

3.1 Coal Petrographic Input in Favour of CTL

3.1.1 Role of Reactive Macerals

Consideration of liquefaction requires that the components of coals are commonly divided into reactive groups (vitrinite and liptinites) and inerts (inertinites and mineral matter). Semifusinite is considered to be partly reactive. It is clear that enhanced conversion will be related to greater amounts of reactives and lower contents of inerts in coal samples. Liptinite group of macerals is rich in hydrogen and are suitable for hydrogenation. Coal with higher liptinite content has higher volatile matter. Generally, higher liptinite content is found within coal with high ash content, and often, the true liptinite content is masked by the mineral matter. During thermal maturation, the kerogen (Type I, II, III and IV) involves hydrogen disproportionate reactions in which the coal and kerogen loses hydrogen in succession. These hydrogen depleted coal and kerogen condenses and aromatizes, eventually to form graphite [28, 29].

Critical importance of liptinites in estimating the potential of coal for oil generation has been assessed, and attempts were made for estimation of the minimum volume of liptinite required for capable of generating oil by many workers [29–31]. A minimum of 15–20% [28, 32, 33] and 20–25% [34] liptinite were estimated for potentiality of coal as oil prone. Assessment of potentiality of hydrocarbon formation of individual liptinite maceral reveals the order as alginite, resinite, cutinite, sporinite and suberinite [35, 36].

The liptinite and vitrinite macerals are transformed to inertinite by virtue of higher temperature and time with increasing rank [37, 38]. The higher temperature and time changes the maceral composition of coal and kerogen thus changing the physical, chemical and optical properties from a sapropelic coal to humic coal and from humic coal towards more humic leading to graphite at last.

Vitrinites typically contain about 70% aromatic carbon atoms in hard brown coals and over 90% in anthracites. Liptinites initially have a very low aromatic content, but in low-rank bituminous coal, 50% carbon atoms are found in aromatic rings. Thereafter, the aromaticity rises rapidly to join that of vitrinite in anthracites. The inertinite maceral group has 90–100% aromatic carbon at all ranks. This invariably supports that hydrogenation is the basic process by which aliphatic-rich liptinite maceral may be derived from more aromatized vitrinite having a large range of aromaticity, while it is quite impossible to revert back from the maximum aromatized inertinite maceral in any way.

Some vitrinite-rich coals have hydrogen content (4.6–5.4 wt%), higher than the normal vitrinites, and show reddish-brown fluorescence under UV light. These hydrogen-rich vitrinites are called perhydrous vitrinite, found particularly in marine-influenced coals with increased wax–resin component [37]. The visible fluorescence of these vitrinites is commonly taken to be a strong indicator of a hydrogen-rich composition when chemical analyses are not available. The maceral composition of perhydrous vitrinite rich and liptinite poor New Zealand coals [39–42] confirms the potentiality of perhydrous vitrinite as an important source of liquid hydrocarbons in oil-prone coals. Similar observation was also noted from Cretaceous coals of the San Juan Basin, New Mexico [43], Gippsland Basin, Australia [44, 45]. In humic coals, high liptinite contents are commonly associated with perhydrous vitrinite compositions [46–48].

3.1.2 Vitrinite Reflectance (VR) and Effect of Its Suppression

Vitrinite reflectance (VR) is the proportion of incident light reflected from a polished vitrinite surface. Vitrinite is a maceral (an organic component) of coal. As vitrinite is heated in the earth during burial, it systematically changes its reflective properties. These changes are caused by increasing condensation of aromatic (carbon ring) structures in the coal matrix [30, 49]. Vitrinite reflection is a standard method for calculating the relative degree of coalification to reveal the rank and maturation that a coal has undergone. Coals with VRo% less than 0.45 lie within the zone of early stage of oil generation, whereas the values of 0.5–0.57% indicate the beginning of commercial oil generation from coal within the diagenetic stage [28].

Suppression of vitrinite reflectance is a common phenomenon in which reduced reflectance values are accounted lower than those produced by low reflecting, non-suppressed vitrinite. Suppressed reflectance values are intermediate between those for liptinite and those for non-suppressed normal vitrinites with hydrogen-poor lignocellulosic structures. Such suppression may occur through the modification of vitrinite structure due to interplay of a number of factors like the high hydrogen contents produced by reducing environment of deposition, impregnation of high bitumen contents produced during early stages of diagenesis along with the formation of perhydrous vitrinites, highly aliphatic components formed due some specific palaeoecological factors or presence of high liptinite content and type of liptinite maceral present in the maceral assemblage [50]. Quantitative assessment of liptinite required for suppression of vitrinite reflectance varies from at least 20% [51] to 35% having 24% resinite component [50]. Suppression effect is primarily attributed to variation in the hydrogen content of vitrinite, while anomalously low reflectance is exhibited by perhydrous vitrinites [52, 53].

3.2 Expulsion of Liquid Hydrocarbon

Extent of expulsion of generated liquid hydrocarbons from coal-bearing sequences is probably the most critical factor in determining whether or not a coal has given rise to liquid petroleum products. Evidences are there for movement of petroleum molecules from their structural site of origin and redistribution of generated bitumen in the form of exsudatinite in fractures and fluorescing cell fillings in macerals within coal seams [54, 55]. Poor expulsion is attributed to the adsorption of oil on the organic macromolecule.

3.3 CTL Processes

Coal liquefaction is a general term referring to a family of processes for producing liquid fuels from coal. Specific liquefaction technologies generally fall into two categories: direct (DCL) and indirect liquefaction (ICL) processes. Indirect liquefaction processes generally involve gasification of coal to a mixture of carbon monoxide and hydrogen (syngas) and then using a process such as Fischer–Tropsch (FT) to convert the syngas mixture into liquid hydrocarbons. By contrast, direct liquefaction processes convert coal into liquids directly, without the intermediate step of gasification, by breaking down its organic structure with application of solvents or catalysts in a high pressure and temperature environment. Since liquid hydrocarbons generally have a higher hydrogen–carbon molar ratio than coals, either hydrogenation or carbon-rejection processes must be employed in both ICL and DCL technologies. South Africa developed CTL technology at Sasol in the 1950s during an oil blockade, and CTL now plays a vital part in South Africa's national economy, providing over 30% of their fuel demand. DCL works by dissolving the coal in a solvent at high temperature and pressure. This process is highly efficient, but the liquid products require further refining to achieve high-grade fuel characteristics. ICL processes operate in two stages. In the first stage, coal is converted into syngas (a purified mixture of CO and H_2 gas). In the second stage, the syngas is converted into light hydrocarbons using one of three main processes: Fischer–Tropsch synthesis, methanol synthesis with subsequent conversion to gasoline or petrochemicals and methanation. Carbon capture and storage, which is still the subject of much research, would alleviate the environmental impact of carbon dioxide being released into the environment, the main argument against CTL by critics.

3.4 Indian Scenario, Recent Advancement and Future Potential

Permian coal of India is composed of variable proportion of vitrinite and inertinite (17–70%) with subordinate amount of liptinite (<20%) [56]. The average maceral composition of Permian coal of India indicates lower fusible or reactive (vitrinite, exinite and semivitrinite) and higher 'inert' constituents and mineral matter similar to Permian coal of western Australia and South Africa [57]. As a whole in all the Gondwana basins of India, vitrinite content increases upward along the stratigraphic succession. General value of vitrinite content in Karharbari coal seams is <20% (fusic-vitrofusic), in lower Barakar coal seams 25–40%, (trimaceritic), in upper Barakar Member 50–70% and in Raniganj coal seams up to 70–80% (vitric-fusovitric). Coking property of coal is also a function of reactive constituent (vitrinite + liptinite) content. In Indian Gondwana basins, high liptinite value up to 25% is found within the coals of Mahanadi and Son Valley although isolated high values are reported from almost all the coalfields. GSI is having a comprehensive inventory of petrographic composition and proximate and ultimate analytical data of Indian coal, generated through decades through exploration activities of coal. Coal petrographic study of some regional coal seams of Ib-River and Talchir Coalfields of Odisha and Mand-Raigarh Coalfield of Chhattisgarh of Mahanadi Valley Gondwana master basin, so far, has already established their suitability for potentiality of oil generation [58, 59]. They meet the basic petrographic criteria for potentiality of oil generation, i.e. Vitrinite + Liptinite > 60%, vitrinite reflectance value (Ro%) < 0.80 and H/C atomic ratio > 0.75. Along with petrographic character of the coal seams, their thickness and depth of occurrence are also favourable for CTL process.

3.4.1 Talcher Coalfield, Mahanadi Valley Basin

Detailed coal petrographic study carried out in different exploration blocks by GSI revealed that coals from Talcher Coalfield have vitrinite (20–65%), liptinie (12–54%) and inertinite (7–28%), the total reactive contents (vitrinie + liptinite) of most of the coal seams being quite high (up to 74%), which is very much desirable for coal liquefaction process. Occurrence of increased liptinite content may be due to decomposition of tissue cells during fusinitization process and redistribution of the bitumen within the fusinite [58]. Liptinite macerals are composed of sporinite, sporangium, cutinite, resinite, alginate, suberinite and liptodetrinite. Coal seams are of subbituminous to high volatile bituminous 'B' type with VRo% varying from 0.30 to 61. During bituminization, perhydrous vitrinite has also taken an active part to form petroleum-like substances. Depth (208–279 m) and thickness (0.58–47.35 m) of the ten regional Barakar coal seam zones (II to XI) have mineable prospect through suitable CTL technology.

3.4.2 Ib-River Coalfield, Mahanadi Valley Basin

Regionally co-relatable subbituminous R-I and R-III Raniganj coal seam zones were found to exhibit high percentage of vitrinite (38.53–53.92%) group of macerals followed by consistently higher liptinite content (12.92–27.45%) with VRo% ranging from 0.31 to 0.40 [59]. Again sub-bituminous to high volatile bituminous C-type regional Barakar Coal seam zones of Belpahar, Parkhani, Lajkura and Rampur show moderate to high vitrinite (12.48–67.60%) followed by liptinite (12.43–35.92%) with VRo mean% ranging from 0.30 to 0.53 [59]. Total vitrinite present in the coal may act as reactive maceral along with liptinite indicate in the process of coal liquefaction. In general, considerable persistent thickness of the coal seams occurring within 700 m may open up a new vista for the future potentiality inviting modern CTL process.

3.4.3 Mand-Raigarh Coalfield, Son Valley Basin

Sub-bituminous to high volatile bituminous 'C'-type regional Barakar coal seams (Nos. IX, VII, VI, IV, III, II and I) show a moderate to high vitrinite (11.14–72.39%) and liptinite (2.68–11.73%), resinite being most abundant, which show higher value (12.46–25.92%) with VRo ranges from 0.35% to 0.57% [59]. Thickness and depth of occurrence within 650 m of the coal seams render a good prospect for CTL.

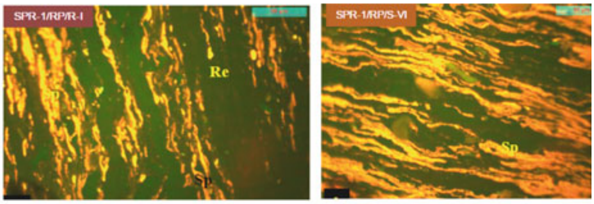
3.4.4 Godavari Coalfield

In the north-western part of the coalfield from Belampalli area, the middle to upper part of thick 'Bottom seam' of high volatile bituminous 'C' type is comprised of vitrinite (25–44%), liptinite (24–32%) and inertinite (11–26%) group of macerals with the Ro mean of 0.50% stage [60]. Petrographic analysis of four economic coal seams (I, II, III and IV) from Ramagundam north-western part of the coalfield indicates that these coals are dominated by inertinite followed by vitrinite and liptinite [61]. The vitrinite group of macerals ranges from 20 to 64% in Seam IIIA, 31 to 57% in Seam II and 24 to 63% in Seam III. The liptinite macerals vary between 10 and 18% in Seam IIIA, 12 and 19% in Seam II, and 10 and 17% in Seam III. In the Koyagudem area of the south–eastern continuity of Mulug Coal belt located in the central part of Godavari Coalfield also reveals favourable petrographic characters for CTL. The 'Queen Seam' occurring between 163.00 m and 185.50 m depth range has shown vitrinite as the dominant maceral group (40–59%), followed by inertinite (21–35%) and liptinite (9–23%). The basal part of Queen seam has dominance of liptinite (33–38%). In general, Ro mean% ranges between 0.48 and 0.64, which shows that this coal seam has reached high volatile bituminous C rank [60]. Domainal enrichment of reactive macerals (>60%) of some selected thick coal seams throughout Godavari Coalfield from NW to SE warrants its introspection for their CTL potentiality.

3.4.5 Lignites of Cambay Basin

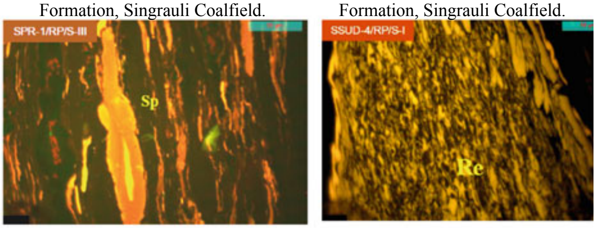
Petrographic study of the three lignite fields, i.e. Vastan, Rajpardi and Tadkeshwar of the Cambay Basin of Gujarat was studied in detail [62]. The lower and upper Vastan lignite seam consists of huminite (58.0–83.1%), followed by liptinite (6.9–16.6%) and inertinite (1–17.9%). In Tadkeshwar upper and lower lignite seam, huminite (52–75.9%) is the dominant maceral group followed by liptinite (6.8–24.4%) and inertinite (1.4–14.8%). Rajpardi lignite comprises of huminite (67.9–80.0%), liptinite (8.7–16.3%), followed by inertinite (2.8–9.3%). The huminite reflectance of the lignites of Cambay Basin shows that Ro mean varies from 0.27% to 0.34% (average 0.31%). All the lignite petrographic data show favourable character for CTL.

Photomicrographs of some Barakar and Raniganj coal seams enriched in different varieties of liptinite macerals from Singrauli Coalfield, Madhya Pradesh; Ib-River Coalfield, Odisha; and Raniganj Coalfield, West Bengal, are given here (Fig. 5a–j). Petrographically, all the coal seams indicate their oil-prone nature [63].



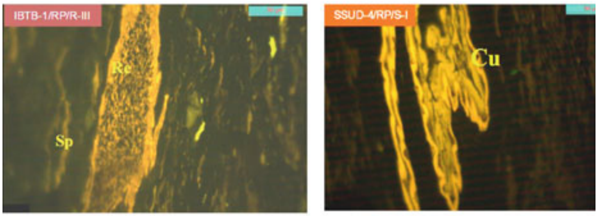
a. Thin elongated soprinites (Sp) and small globules of resinite (Re). Raniganj Formation, Singrauli Coalfield.

b. Liptinite rich bands with thin elongated soprinites (Sp) Barakar Formation, Singrauli Coalfield.



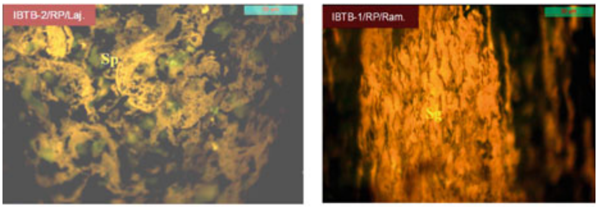
c. Both micro and mega sporinites (Sp), Barakar Formation, Singrauli Coalfield.

d. Thick resinite (Re), Barakar Formation, Singrauli Coalfield.



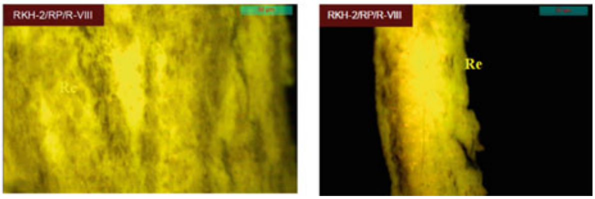
e. Thick band of resinite (Re), with sporinite (Sp), Raniganj Formation, Ib-River Coalfield.

f. Cutinite (Cu) in association with collodetrinite, Raniganj Formation, Ib-River Coalfield.



g. Sporinite (Sp) clusters, Barakar Formation, Ib-River Coalfield.

h. Large sporangium, Barakar Formation, Ib-River Coalfield.



i. Thick band of resinite (Re), Raniganj Formation, Raniganj Coalfield.

j. Thick band of resinite (Re), Barakar Formation, Raniganj Coalfield.

Fig. 5 a–j Photomicrographs of some Barakar and Raniganj coal seams from different coalfields enriched in different varieties of liptinite macerals

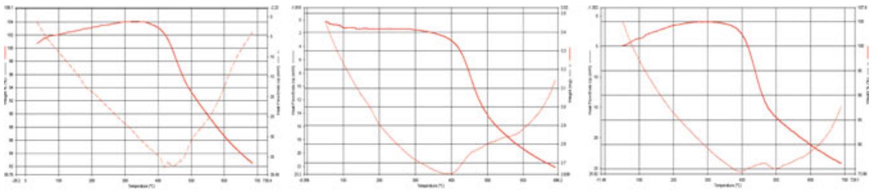
3.5 Recent Advances

In this context, a thorough petrographic scan of the low- to medium-rank Gondawana coal and Tertiary lignite and low-rank Tertiary coal of North East India is required with special reference to suitability for CTL. Multiparametric approach including petrography, thermal, ultimate and proximate analyses, spectroscopy using UV and IR may be used to identify the oil-prone nature. Coal oxidation at moderate temperatures ($\leq$400 °C) is a complicated process affected by several factors, including coal compositions, temperature and oxygen concentration in the environment and alters the coal properties as fuel, thereby making pronounced impact on its subsequent utilization [64, 65]. Recent researches are going on in this direction with different spectroscopic techniques on coals and their soluble extracts to understand the macromolecular structures, which is essential for evaluate the oil producing and expelling potentiality of them.

3.5.1 TGA-DTA Analysis

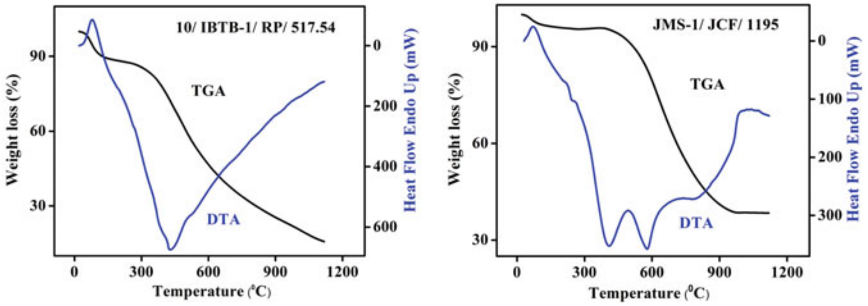
Thermogravimetric analysis (TGA) and derivative thermogravimetry analysis (DTA) studies are useful tools for the determination of combustion characteristics (e.g. peak temperature, burnout temperature and activation energy) of coals during heating in a controlled system [66–68]. The procedure is strongly affected by particle size, ash content, sample amount, heating rate and gas flow rate [66]. High heating rates will cause simultaneous evolution and ignition of volatiles, whereas with low heating rates devolatilization will occur prior to ignition and combustion. The burning profile of coal can be an instrumental analysis in distinguishing between different coals. TGA curves show the change in weight of coal as a function of temperature and corresponding DTA curves shows the thermal events like exothermic and endothermic reaction, decomposition, melting, crystallization and change in crystal structure, etc. Analysis have been carried out for low-rank liptinite-rich non-coking coal of Ib-River Coalfield, Odisha, high-rank coking coal of Jharia Coalfield, Jharkhand, non-coking coal of Raniganj seams of Raniganj Coalfield and Tertiary coal of Meghalaya which gives some important inference regarding the energy release and decomposition behaviour of coal under a programmed and controlled environment (Fig. 6).

The thermal properties of coal strongly depend on coal rank. The ignition temperature of coals increases with decreasing volatile matter content. The ignition temperatures of maceral groups increase in the order of liptinite–vitrinite–inertinite [69]. Some recent studies have also shown that the combustion behaviour of coal cannot be merely correlated to petrographic composition. Maceral interactions can occur, and depending on the rank of coal, liptinite has a more important role in the maceral interaction [67]. The rank or parameter comprising rank and petrographic composition has more effect on combustion behaviour than merely maceral composition does [70, 71].



R1 and R5 Raniganj coal seams, Raniganj Coalfield

Tertiary sulphur-rich non-coking coal, Meghalaya



Non-coking Lajkura coal seam of Barakar Formation, Ib-River Coalfield

Coking coal seam of Barakar Formation, Jharia Coalfield

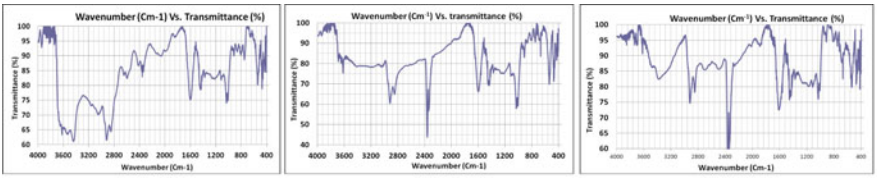
Fig. 6 TGA-DTA profiles of coal seams from different coalfields

3.5.2 FTIR Spectral Analysis

FTIR spectra of the non-coking Gondwana coals from R-I and R-V Raniganj seams of Raniganj Coalfield, West Bengal, Lajkura and Parkhani seams of Barakar Formation of Ib-River Coalfield, Odisha and Tertiary coal from Meghalaya indicate distinctly different signals indicating different macromolecular structures (Fig. 3) The peak ranging from 2800 to 3700 cm^{-1} is due to the major water and presence of C–H stretching (2800–3100 cm^{-1}) and C–H bending (700–900 cm^{-1}) of hydrocarbon components. A distinct peak around 1600 cm^{-1} region is due to the C=C ring stretch mode arising from irregular aromatic units as polycyclic aromatic hydrocarbons (PAH) containing ortho-substituted end rings or substituted with aliphatic chains (Fig. 7). Enriched aliphatic signatures of coals from Ib-River Coalfield indicate its oil-prone nature.

3.5.3 Ultraviolet (UV) Visible Spectroscopy

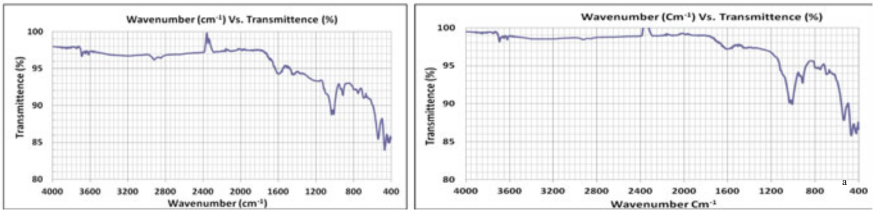
Analysis of UV-visible spectra of the water-soluble condensate of Lajkura coal seam of Barakar Formation of Ib-River Coalfield derived from destructive distillation up to 160 °C reveals that UV excitation enhances the visualization of the



R1 coal seam of Raniganj Formation, Raniganj Coalfield

R5 coal seam of Raniganj Formation, Raniganj Coalfield

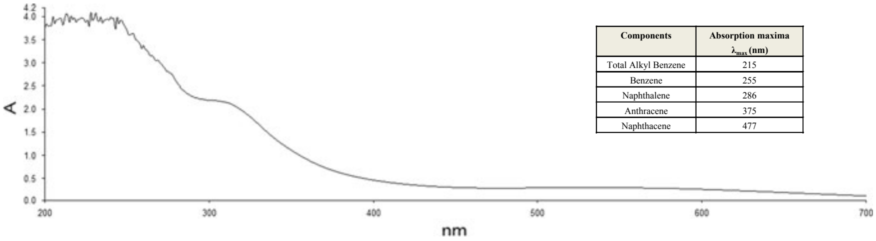
Tertiary coal seam, Meghalaya



Lajkura coal seam of Barakar Formation, Ib-River Coalfield

Parkhani coal seam of Barakar Formation, Ib-River Coalfield

Fig. 7 FTIR spectra of coal samples from different seams of Gondwana and Tertiary coalfields



Components	Absorption maxima λ_{max} (nm)
Total Alkyl Benzene	215
Benzene	255
Naphthalene	286
Anthracene	375
Naphthacene	477

Fig. 8 The wavelength (nm) versus absorption (A) spectra of condensate dissolved in water

structures of chromophores, the light incorporating group of the molecules. The absorption spectra of condensate clearly demonstrate the presence of dominantly aromatic ring hydrocarbon of composition of alkyl benzene, benzene and naphthalene with lesser presence of higher ring polycyclic aromatic hydrocarbons (Fig. 8).

3.5.4 Ultraviolet (UV) Fluorescence Spectroscopy

Fluorescence spectroscopy, in the form of steady-state emission (SSE) and synchronous fluorescence scanning (SFS), is a diagnostic tool for investigating polycyclic aromatic hydrocarbon (PAH) structures in terms of the size of the fused aromatic ring (FAR) present in the condensate sample. It has been found from the

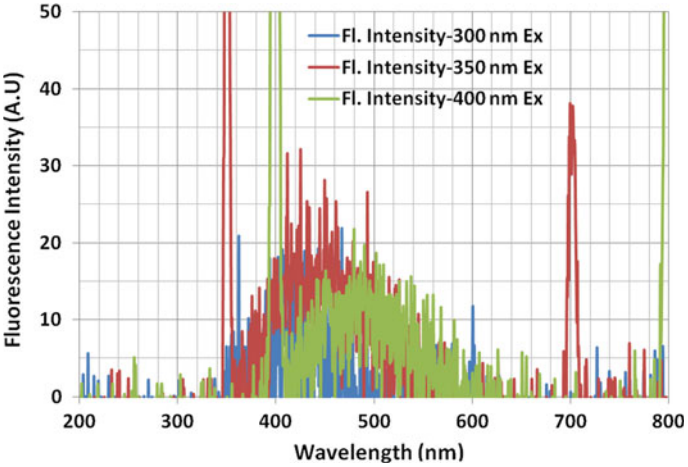


Fig. 9 Steady-state emission spectra (at 300 nm, 350 nm and 400 nm excitation wavelength) of condensate dissolved in water

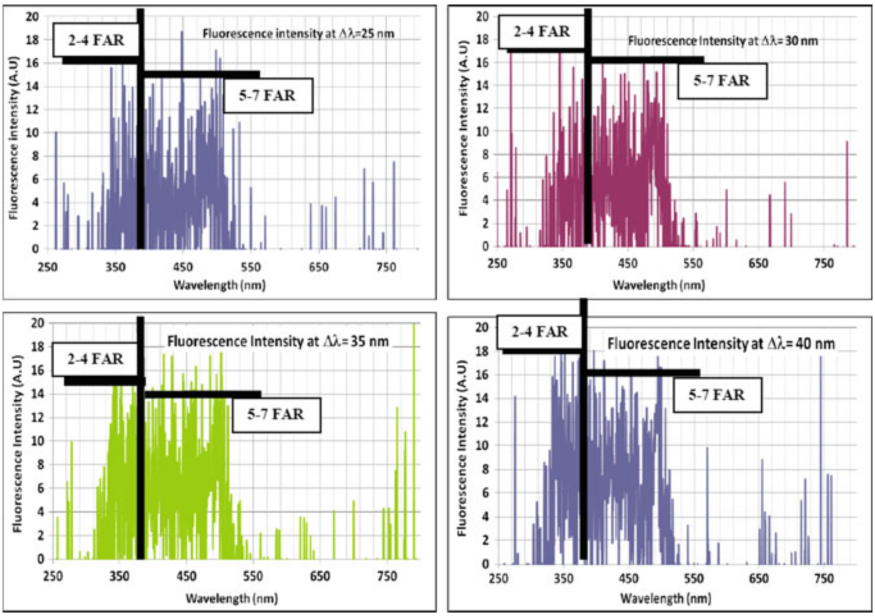


Fig. 10 Synchronous fluorescence spectra of the water dissolved condensate at **a** $\Delta\lambda = 25$ nm, **b** $\Delta\lambda = 30$ nm, **c** $\Delta\lambda = 35$ nm and **d** $\Delta\lambda = 40$

above figure that the SSE spectra at different excitation wavelengths are exhibiting a characteristic broad range of emission pattern from 360 to 650 nm. This observation can be correlated with the fact that the condensate sample contains high molecular weight polycyclic aromatic ring structures, i.e. asphaltenes (Fig. 9).

For the detailed investigation of the ring size (fused aromatic ring, FAR) associated with the high molecular PAH structures in the condensate, SFS spectra of the water-soluble condensate are captured which shows the presence of both small (2–4) FAR and larger (5–7) FAR (Fig. 10).

4 Conclusions

Studies undertaken so far from coal and lignite of non-coking and coking coal of both Raniganj and Barakar formations of Gondwana coalfields and Tertiary Coalfields of North-eastern region as well as lignites from west and coast lignite fields, unravel the intrinsic potentiality of some of them for unconventional greenfield energies like gas and oil. A huge data accrued through exploratory drilling in different coal and lignite fields are available, which are to be introspected in detail to unlock their potentiality. Favourable factors with respect to physic-mechanical, chemical, structural and petrographical characters are present for Indian solid fossil fuel, which may be utilized with advanced site-specific and composition-specific technology in near future. Maximum thrust is to be given on the different spectroscopic studies of coal and lignite to achieve the fundamental knowledge of their macromolecular structure so that they may be used more effectively for specific purposes of UCG or CTL. Understanding the size of various polyaromatic components released during heating of coal will help to design customized systems for coal gasification/liquefaction as well as residual materials which could be used as carbon feedstock. Understanding of this specific knowledge of aromatic components in coal has direct impact to determine the internal macromolecular structures to ascertain their suitability for coal utilization process.

Acknowledgements SB thankfully acknowledges Director General, GSI, for giving opportunity to represent GSI in the 'one-day conference' on 'Macromolecular characterization of coal and hydrocarbon components for future' jointly organized by School of Petroleum Technology, Pandit Deendayal Petroleum University (PDPU), Gujarat, with S. N. Bose National Centre for Basic Sciences, Kolkata, on 125th Birth Anniversary of Prof. Satyendra Nath Bose held on 19th November, 2018, in PDPU Campus, Gandhinagar. The present paper is an outcome of the deliberation of SB in this conference. He also thankfully acknowledges the support of Petroleum Engineering Laboratory of PDPU for the spectroscopic analysis and characterization of data. Data generated by GSI, which have been used in this paper are taken from the different unpublished technical reports of investigations carried out as approved FSP of GSI.

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