

Coaly source rocks of NW Borneo: role of suberinite and bituminite in oil generation and expulsion

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Abstract: The coaly source rocks currently investigated are of Tertiary age, collected from areas of Bintulu, Sarawak and Labuan Island, East Malaysia, and Brunei-Muara, Brunei Darussalam. Much of the petroleum in the Southeast Asian region was generated from organic matter of higher plant origin deposited within a lower delta plain to prodelta setting. However, the specific nature and origin (other than being 'higher plant') of the organic matter, has not been identified. In this study, the maceral suberinite and, the associated maceral, bituminite have been recognised as the most oil-prone macerals. It was petrographically observed that subsequent to hydrocarbon generation, the suberinite-phlobaphinite framework breaks down, giving rise to liptodetrinite, fluorinite, vitrodetrinite and exsudatinitite. Accumulation of disordered or disintegrated suberinitic constituents subsequently formed bituminite. These suberinitic-rich coals appear to be associated with mangrove precursors prominently within paralic and shallow marine depositional settings. It was also noted that these hydrogen-rich constituents may have been transported into relatively deeper marine environments as suggested by the presence of suberinite in turbiditic sediments from Labuan Island.

Keywords: oil-prone coals, coaly sediments, suberinite, bituminite, exsudatinitite, maceral transformation, hydrocarbon generation and expulsion, mangrove-derived constituents, biomarkers.

INTRODUCTION

Tertiary coals of Southeast Asia are known to be effective source rocks in the prolific gas- and oil-producing basins of the region (e.g. Todd *et al.*, 1997; Mazlan and Abolins, 1999). However, the nature and origin (other than the generic 'higher plant') of the organic matter, has not been identified. The depositional setting of these oil-prone coals is generally considered to be within a broad setting of lower delta plain to prodelta setting. The aim of this study is to demonstrate that suberinite, and its associated macerals, are the main oil-prone macerals of NW Borneo coals, and to discuss their origin, occurrence and depositional setting. The coals and coaly sediments under current investigation are outcrop samples of Tertiary age, collected from areas around Bintulu and Labuan Island, East Malaysia, and from the Brunei-Muara District, Brunei Darussalam (Fig. 1).

In this study, petrological characteristics, in combination with organic geochemical parameters, commonly considered when assessing the origin of source material for oils will be presented. About fifty coal/carbargillite samples and two carbonaceous shale/siltstone were evaluated by organic petrological methods. A number of these samples were subjected to organic geochemical

analyses. Petrographic studies were performed in plane-polarised reflected light using a Leica DMRX photometry microscope equipped with fluorescence illuminators. Microscopical examination was carried out using oil immersion objectives under normal reflected 'white' light and under blue light excitation. Vitrinite reflectance measurements were principally carried out in 'white' light (at 546 nm) using an oil immersion objective. Microscopical identification of specific organic matter types and their origins are highlighted as these are most appropriate in complimenting the organic geochemical data. The geochemical analyses carried out include bitumen extraction, gas chromatography-mass spectrometry (GC-MS) and pyrolysis (Py)-GC.

ORIGIN OF SUBERINITE

Suberinite is derived from "suberin" which is found in corkified cell walls which occur mainly in barks, and also at the surfaces of roots, on stems and on fruits (Teichmüller *et al.*, 1998). Suberin consists of glycerine esters of high molecular weight, unsaturated and saturated fatty acids and oxy-fatty acids (Kölattukudy, 1980). Cork cells consist of layers of cellulose, lignified cellulose as well as suberin. Primitive plants, which were predominant

in the Carboniferous, did not form cork. Thus, suberinite is a maceral that is almost exclusively found in Tertiary coals and a few Mesozoic coals (Teichmüller, 1982).

GEOLOGICAL SETTING OF NW BORNEO

The major basins of the Northwest Borneo region include the Sarawak Basin, the northwest Sabah Basin and the Baram/Champion Delta (Fig. 1) which developed on a broad shelfal area after the deformation and uplift of the Rajang Group (Haile, 1969). Erosion of this mountainous terrain provided the clastic supply for these basins. Mainly coarse clastics were deposited in the Late Eocene to Oligocene while predominantly carbonates, argillaceous sediments and sand bodies were deposited in Middle Eocene to Miocene times. The early Miocene limestones were overlain by shales and marls of the Sibuti Formation and Setap Shale. These predominantly argillaceous marine sediments mark a major relative rise in sea level towards the end of early Miocene, during which the limestone reef complexes were drowned. These Tertiary shelfal sediments were gently folded with a NNW to SSE trending axis during the middle Miocene time and were subsequently overlain unconformably by the Neogene Belait Group (Haile, 1969). Pliocene-Pleistocene folding and uplift affected all

the older stratigraphic successions and eventually exposed them.

The provinces of NW Borneo as shown in Figure 1 are tectono-stratigraphic subdivisions that are essentially based on structural characteristics and sedimentary facies. Faults and/or plate boundaries that are interpreted as significant crustal discontinuities commonly define the province boundaries (James, 1984). Such a significant boundary is the West Baram Line which is interpreted as a significant crustal discontinuity separating the Luconia Carbonate Province from the coeval deltaic clastic province of the Baram Delta. In this study the coal samples from Brunei occur within the Champion Delta system which bounds the Baram Delta on the east and extends inland of Negara Brunei Darussalam, Labuan Island and Klias Peninsula of Sabah. The Baram Delta (Late Miocene-Quaternary) and the Champion Delta (Middle-Late Miocene) prograded rapidly during the Late Neogene time and developed over the foundation formed by the uplifted and deeply eroded Crocker-Rajang accretionary complex (Koopman, 1996). Sediment supply to the Baram/Champion deltas is predominantly from this Crocker-Rajang hinterland.

In the Bintulu area, the coal seams are found interbedded within the sandstone, shale and clay of the Nyalau Formation. The formation is weakly to

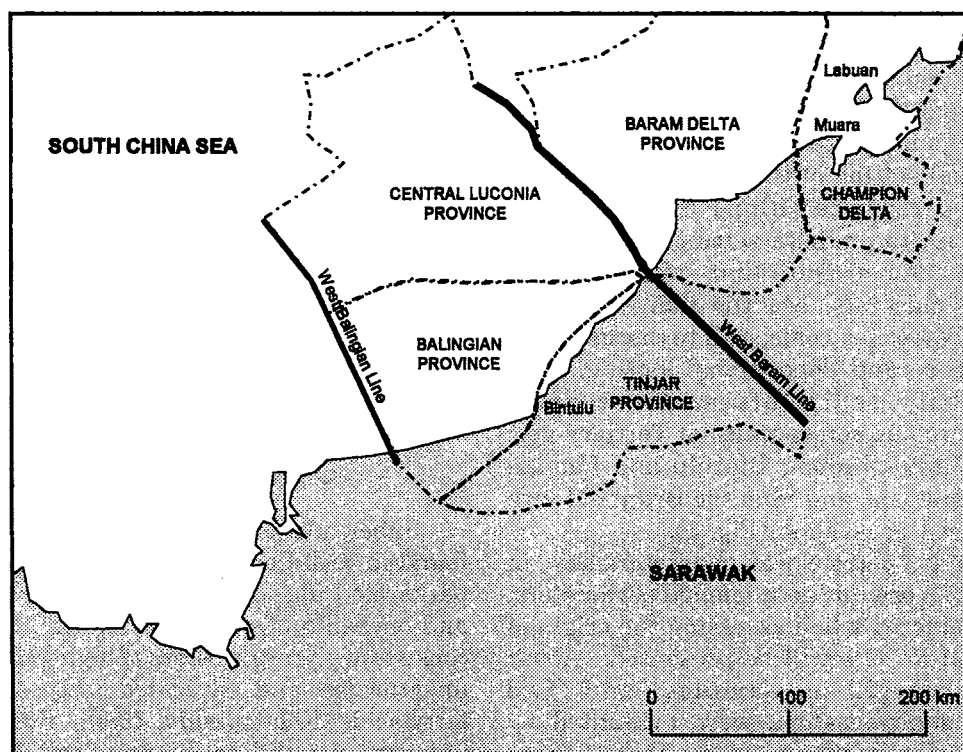


Figure 1. Simplified stratigraphic subdivision of NW Borneo provinces and location of the study areas (modified from Mazlan B. Hj. Madon, 1999).

moderately folded and faulting is common. The Nyalau Formation (Oligocene to early Miocene) is predominantly arenaceous consisting of alternating thin and thick sandstone beds with subordinate shales that increase towards the top. The depositional facies, as identified by Bait and Asut (1991) represent shallow marine, tidal and coastal plain deposits.

The Brunei coals studied here are from the Middle Miocene Belait Formation that was deposited in a lower flood plain under tidal influence (Tate, 1976; Yap, 1996). The samples from Labuan are from Tg. Kiamsam of the East Kiamsam Sandstone slump unit (Lee, 1977). The unit was mapped as Temburong Formation (Oligocene to Lower Miocene) by Wilson (1964) who considered it, in part, a time equivalent to the West Crocker Formation of Padas Valley area, Sabah which was deposited mainly by turbidity currents and contain a mixed flysch-type deposit.

HYDROCARBON-BEARING COAL SEQUENCES

The prospective hydrocarbon accumulations of Northwest Borneo, as with most Southeast Asian basins, are of Tertiary age and are commonly associated with coal-bearing sequences. A thick sedimentary sequence of up to 5,000 m of the Upper Tertiary sediments is the main hydrocarbon-producing interval of the NW margin of Borneo (James, 1984). The sedimentary environments range from fluvial-deltaic, estuarine, marine shelf and turbidite. The petroleum provinces of Balingian and the Baram Delta both belong to paralic lower delta plain to prodelta settings although lacustrine sources have been postulated (Todd *et al.*, 1997). As reported by these workers, the paralic oil-prone source rocks include both coals and carbonaceous shales/mudstones containing higher land plant-derived organic matter.

The Balingian Province is bounded to the north by the central Luconia Province, to the west by the West Balingian Line and to the south by the Tinjar Province (Fig. 1). The southern most part of the province extends into onshore Sarawak while the eastern boarder is bounded by the West Baram

Line. The greater part of the province occurs in the offshore area of Sarawak. It is an actively producing petroleum province with the first discovery being in Temana Field in 1962. Hydrocarbons occur in Upper Oligocene to Lower Miocene siliciclastic sediments. The main source rocks, especially within west Balingian, are coals of a lower coastal plain setting (Mazlan and Abolins, 1999). Towards the east, the source rocks are reported to become increasingly marine-influenced and increasingly gas-prone. For a thorough review on the exploration history and the petroleum system of this province, readers can refer to the workers mentioned above.

PETROGRAPHIC DATA

A summary of the maceral composition for about fifty coals and coaly sediments from Bintulu and Brunei-Muara analysed in this study is shown in Table 1. A reasonably well preserved suberinite associated with framboidal pyrite is observed in the carbonaceous shaly siltstones from Kiamsan Peninsula, Labuan Island (Figs. 2 and 3). The level of thermal maturation attained by these sediments, as indicated by vitrinite reflectance, is about 0.6%Ro. In general, based on vitrinite reflectance, the coals under current investigation are of sub-bituminous rank (C - A), ranging from about 0.4 to 0.7%Ro. This range of thermal maturity would generally be regarded as immature to early mature for oil-generation (e.g. Tissot and Welte, 1984). Coals from southern Bintulu are of relatively higher thermal maturity compared to those from northern area, whilst coals of the Brunei-Muara are least mature among the analysed samples.

The dominant maceral in all of the samples analysed is vitrinite. Exinite/liptinite content is high, particularly in the Brunei-Muara and North Bintulu coals in which suberinite and bituminite together make up about half the total liptinite content. Other liptinites such as sporinite, resinite and cutinite are fairly common in most samples. Exsudatinites is commonly observed to be associated with bituminite, and is most prominent in relatively lower thermal maturity samples (0.45–0.55%Ro) in northern Bintulu and Brunei-Muara areas. On average, coals from northern Bintulu are relatively

Table 1. Summary of vitrinite reflectance (%Ro) and maceral composition.

| Locality | %Ro (random) | Vitrinite (%volume) | Liptinite (%volume) | Inertinite (%volume) | Mineral Matter |
|---------------|--------------|---------------------|---------------------|----------------------|----------------|
| Brunei-Muara | 0.40–0.50 | 50 | 35 | 5 | 10 |
| North Bintulu | 0.45–0.55 | 50 | 30 | 5 | 15 |
| South Bintulu | 0.55–0.70 | 70 | 15 | 5 | 10 |

richer in liptinite content compared to those from southern Bintulu.

GEOCHEMICAL DATA

Organic geochemical evaluation performed on those coals rich in suberinite and/or bituminite yielded reasonably high HI values of about 300 mgHC/gTOC (e.g. Wan Hasiah, 1999a). Saturated hydrocarbon fractions of these coals are dominated by higher molecular weight n-alkanes and display a waxy appearance (Fig. 11). The S2 Py-GC pyrogram of a suberinite-rich coal is dominated by n-alkane/alkene doublets as well as aromatic components (Fig. 12), thus supporting the oil-prone nature of suberinite and its associated macerals, in particular bituminite and liptodetrinite. Based on petrographic and geochemical data presented here,

these coals would be considered to possess good oil-generating potential.

Examples of biomarker distributions within the aliphatic fraction of the extracted samples from Brunei-Muara and Labuan are as shown in Figure 13 and Figure 14 respectively. The Bintulu biomarker distributions are not shown here as they have been previously discussed (see Wan Hasiah, 1999a). As was observed within the Bintulu samples, the m/z 191 chromatograms for the samples shown here are also dominated by C₂₉, C₃₀ and C₃₁ αβ hopanes. In most immature samples i.e. from Brunei-Muara, the C₃₁ αβ hopanes (22R) is the most dominant peak. In the relatively mature samples i.e. from Bintulu and Labuan, the presence of the 18α(H)-oleanane (peak 'Ol') is most apparent compared to the relatively lower maturity coals from Brunei-Muara.

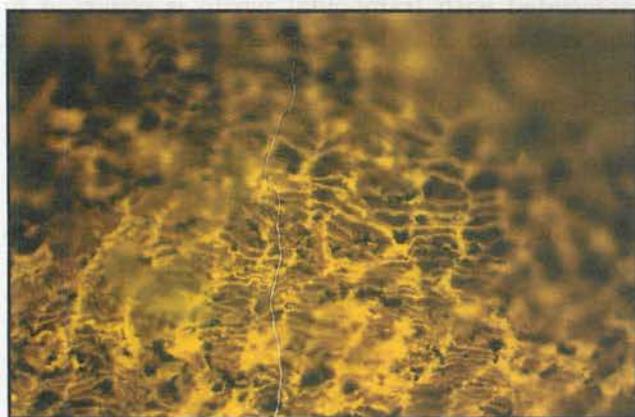


Figure 2. A well-preserved suberinite maceral associated with framboidal pyrite in a coaly sediment of Kiamsam Peninsula, Labuan; blue light excitation, oil immersion, field width = 0.2 mm.

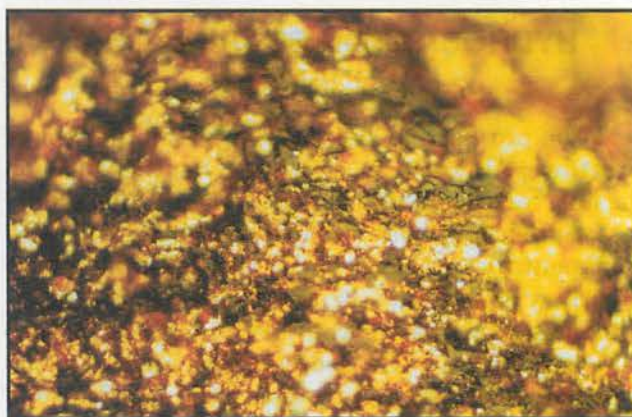


Figure 3. Same view as Figure 2 under reflected white light showing the prominent presence of framboidal pyrite.

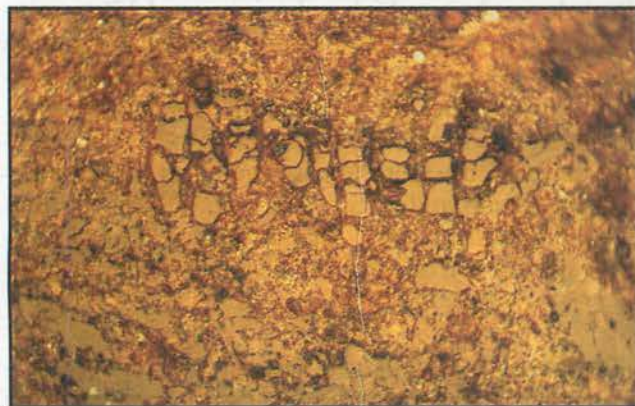


Figure 4. Disintegration of the suberinite-phlobaphinite framework giving rise to vitrodetrinite and associated with heavy oil (bitumen) staining. The dispersed collinite may subsequently coagulate to form collodetrinite (desmocollinite); reflected light, oil immersion, field width = 0.2 mm.



Figure 5. The remnants of the suberinitic cell wall can be distinctly observed under blue light excitation and are associated with brightly fluorescing fluorinite and/or liptodetrinite (same view as Figure 4).

DISCUSSION

Oil-prone macerals

Earlier work (e.g. Khorasani, 1987; Khorasani and Michelsen, 1991) showed suberinite to undergo severe physicochemical alteration to generate substantial amounts of highly waxy hydrocarbons in lignites and sub-bituminous coals. More recent work (e.g. Wan Hasiah 1997 and 1999b), mostly based on low rank coals around Merit-Pila, Bintulu and Miri areas in Sarawak, support these findings and recognised suberinite and the associated maceral bituminite (e.g. Wan Hasiah 1999a; Wan Hasiah 2001b) as the most oil-prone macerals within the studied region.

Transformation of macerals

Under the microscope, the liquid hydrocarbon

generated occurs in the form of oil haze and oil globules, while solid hydrocarbon occurs in the form of exsudatinite which was seemingly soft and mobile when exuded from the precursor material. The texture of the coal fabric and the association of the macerals present are expected to govern the mode of hydrocarbon generation and the expulsion pathways (Wan Hasiah, 2000).

As could be observed petrographically, subsequent to hydrocarbon generation, the suberinite-phlobaphinite framework breaks down, giving rise to liptodetrinite, fluorinite, vitrodetrinite and exsudatinite (Figs. 4 and 5). Accumulation of disordered or disintegrated suberinitic constituents subsequently formed bituminite (Fig. 6). Thus, bituminite (as defined by ICCP, 1975) is recognised here as a secondary maceral (an intermediate stage) which represent a transformation product from the precursor maceral suberinite during hydrocarbon



Figure 6. Accumulation of disordered suberinite forms bituminite (dark brown amorphous mass) seen here surrounding phlobaphinite bodies that have softened and swollen as a consequence of plastic deformation; reflected light, oil immersion, field width = 0.2 mm.



Figure 7. Exsudatinite exerting pressure upon the release of hydrocarbon material forming a network that caused the coal to fracture; blue light excitation, field width = 0.2 mm.

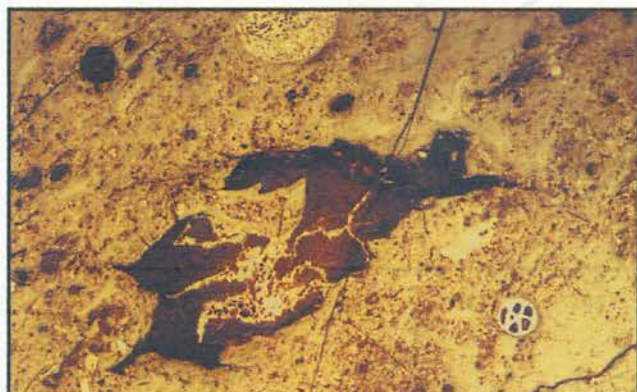


Figure 8. Very low reflecting bituminite closely associated with high reflecting inertinitic veinlets. The 'plastic' nature of these inertinites (that bear great resemblance to fusinite and inertodetrinite) suggest that they are disproportionation products of hydrocarbon generation; reflected light, oil immersion, field width = 0.5 mm.

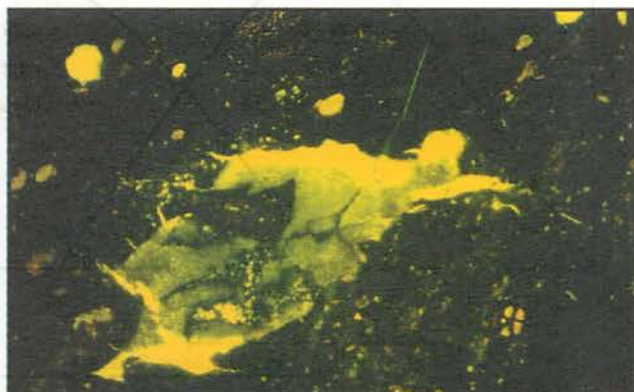


Figure 9. Bituminite commonly shows a dull fluorescence under blue light excitation but fluoresces intense yellow and takes up the form of exsudatinite, oil globules or oil haze when associated with hydrocarbon generation (same view as Figure 8).

generation.

Simultaneous with the transformation of liptinite macerals into liquid hydrocarbon, the maceral vitrinite also undergoes some transformation as a consequence of hydrocarbon generation. The maceral phlobaphinite undergoes plastic deformation (Fig. 6), it softens and subsequently coagulates to form collodetrinite (desmocollinite) that forms much of the coal fabric. When associated with intense hydrocarbon generation, oil globules and gaseous vesicles can form within the phlobaphinitic cells and subsequently they gain higher reflectance. Once solidified, the cells appear very similar to fusinite, macrinite and sclerotinite in morphology and reflectance (Wan Hasiah 2001b). Formation of micrinite and other inertinite macerals appear to be closely associated with that of bituminite (Figs. 8 and 9), occurring mostly as a residual product upon disproportionation of the hydrogen-rich components. It appears that a whole range of maceral arises in association with hydrocarbon generation. Based on these petrographic features, Wan Hasiah (2001b) suggested oil generating coals as a dynamic medium in which macerals are newly created, deformed, disintegrated and thermally decomposed. Subsequently, this would govern the

not just the chemical structure but would impact on the physical character of the coals, such as the development of porosity and permeability, thus bare significant consequences to the capacities and timing of hydrocarbon generation and expulsion.

Hydrocarbon generation and expulsion

The capability of coal to expel the liquid hydrocarbon generated is often regarded as being the most critical criteria when assessing oil-generation from coal (e.g. Scott and Fleet, 1994). Factors related to expulsion of oil from coal have been recently reviewed by Wilkins and George (2002) who acknowledged the difficulties in determining the factors involved, in particular, the expulsion efficiency. It seems that, whether determination was via simulation experiment (e.g. Monthieux and Landais, 1987; Noble *et al.*, 1991) or simply by means of microscopical examination (e.g. Cook and Struckmeyer, 1986; Wan Hasiah, 1999a and 2000; Wan Hasiah and Ouzani 2001) the assessment of expulsion of liquid hydrocarbon from coal is mostly inferred. An inferred timing and factors that govern the expulsion of hydrocarbon in relation to oil generation for the NW Borneo coals is shown in Figure 10. The ease of expulsion (i.e. primary migration) appears to be dependent upon

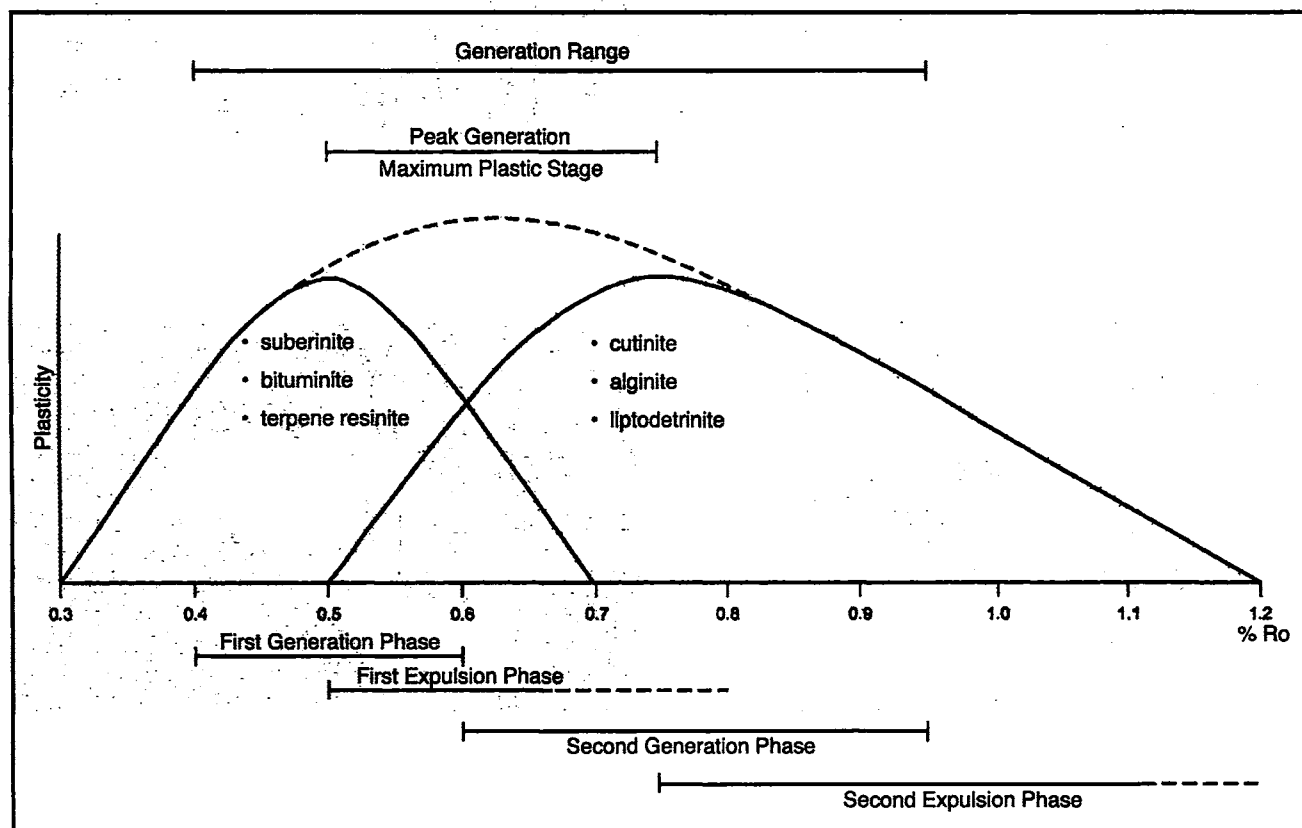


Figure 10. A schematic representation showing the relationship between hydrocarbon generation, expulsion and plasticity (after Wan Hasiah and Ouzani, 2001).

the extent of fracturing within the coal fabric and the development of exsudatinite crack network. Based on previous and the present study of the NW Borneo coals by the current author, exsudatinite is envisaged as hydraulic fractures caused by expulsion of hydrocarbon fluids which is believed to play important role in both hydrocarbon generation and primary migration. It should be noted that upon extensive fracturing, the exsudatinite provides open fractures which are filled by free hydrocarbons as observed petrographically e.g. in the form of oil haze (Fig. 7).

For the coals analysed in this study, hydrocarbon material directly associated with suberinite and bituminite began to generate at about 0.4%Ro and was exhausted by about 0.7%Ro. This early generated material is believed to play a significant role in saturating the source rock pore system and deactivating adsorptive sites as initially proposed by Horsfield *et al.* (1988). Upon reaching a saturation threshold, materials that are expelled should be able to leave the coaly source rock such as via exsudatinite crack networks (e.g. Wan Hasiah, 1999a) without being held back within the pore systems.

Incorporation of remnant suberinitic material within the vitrinite fabric, however, is still apparent in higher maturity coals of about 1.0%Ro (Ouzani and Wan Hasiah, 1998) suggesting that suberinite-derived components, such as fluorinite/liptodetrinite, possess good oil generating potential and expelled their hydrocarbon components at a relatively higher maturation level of between 0.7–1.0%Ro. Inclusions of hydrocarbon generating material and suberinitic components appear to be responsible for the perhydrous nature of the coaly source rocks and for the fluorescence phenomena of the vitrinitic coal fabric.

Biomarker distributions

Oils of West Balingian Province are mainly waxy and show typical biomarker distributions characteristic of a lower coastal plain with significant input of higher plant material as suggested by a high 18 α -oleanane content, a moderate to high bicadinane content and a C₂₉ sterane preference over C₂₇ and/or C₂₈ steranes. In support, the pristane/phytane (Pr/Ph) ratio is high, commonly exceeding 5.0, Pr/nC₁₇ ratio of between 1–1.5 or greater, a high hopane/sterane ratio, a low concentration of diasterane and a moderate to high Tm/Ts ratio (Mazlan and Abolins, 1999). These biomarker distributions are typical of terrestrially-derived oils, specifically coal-sourced, not just for West Balingian oils but widespread within Southeast Asian region as reported by Robinson (1987) and Todd *et al.* (1997). Coal extracts from

the Nyalau Formation, onshore Sarawak show biomarker distributions similar to those offshore (stratigraphic equivalent) of West Balingin Province (Wan Hasiah Abdullah, 1999a). These similarities are most distinct in extracts of coals possessing %Ro of about 0.5–0.6%. The extracts of relatively higher maturity samples (>0.7%Ro) from South Bintulu contain relatively lower oleanane concentration, while extracts of immature coals (about 0.3–0.4%Ro) from Brunei-Muara are lacking in oleanane (Fig. 13).

Origin and depositional setting

Suberinite is known to originate from suberin-impregnated cell walls of cork tissue and occurs mainly in bark but may also be found in the stems and roots of woody and herbaceous plants (see earlier section). *Rhizophora* sp., in particular, contains abundant suberinitic and the associated phlobaphinitic cell fillings (Teichmüller, 1982). It is also known that suberin and subereous components are waxy constituents, considered to be predominantly polymeric containing polyesters and aromatics (Kölattukudy, 1980). Extracts of *Rhizophora* peat were shown to be dominated by higher molecular weight n-alkanes with traces of olean-12-ene present (Dehmer, 1995). Biomarker components, such as oleanane, that commonly designate fluvio-deltaic facies (e.g. Robinson, 1987) are also common in coal extracts from the Nyalau Formation (Wan Hasiah, 1999a) and oils of the Balingian province (Mazlan and Abolins, 1999). The generation of fluvio-deltaic oils has been associated with terrestrially-derived organic matter of mangrove origin (e.g. Brown, 1989, Wan Hasiah 2001a). Mangrove pollen grains, particularly of Rhizophoraceae, are known to have been widely distributed within the Sunda region since early Tertiary times (Morley, 2000) and at Present time (Lambiase *et al.*, 2000). As mangrove paleo-belts are widespread between latitudes 30°N and 30°S, it seems apparent that oil pools within these regions, particularly Southeast Asia, are sourced from mangrove-derived coaly constituents of which suberinite and its associated maceral bituminite are the dominant components.

It is interesting to note that suberinite-bearing coaly constituents are present in deep marine sediments of Temburong Formation of Wilson (1964) from Labuan Island (Fig. 2). This suggests that the incorporation and transportation of the hydrogen-rich constituents such as suberinite is probable within turbiditic sediments. Being deposited in deep, bottom conditions of deposition, these oil-prone constituents appear to have not only survived the transportation and redeposition process but also appear to have retained or enhanced

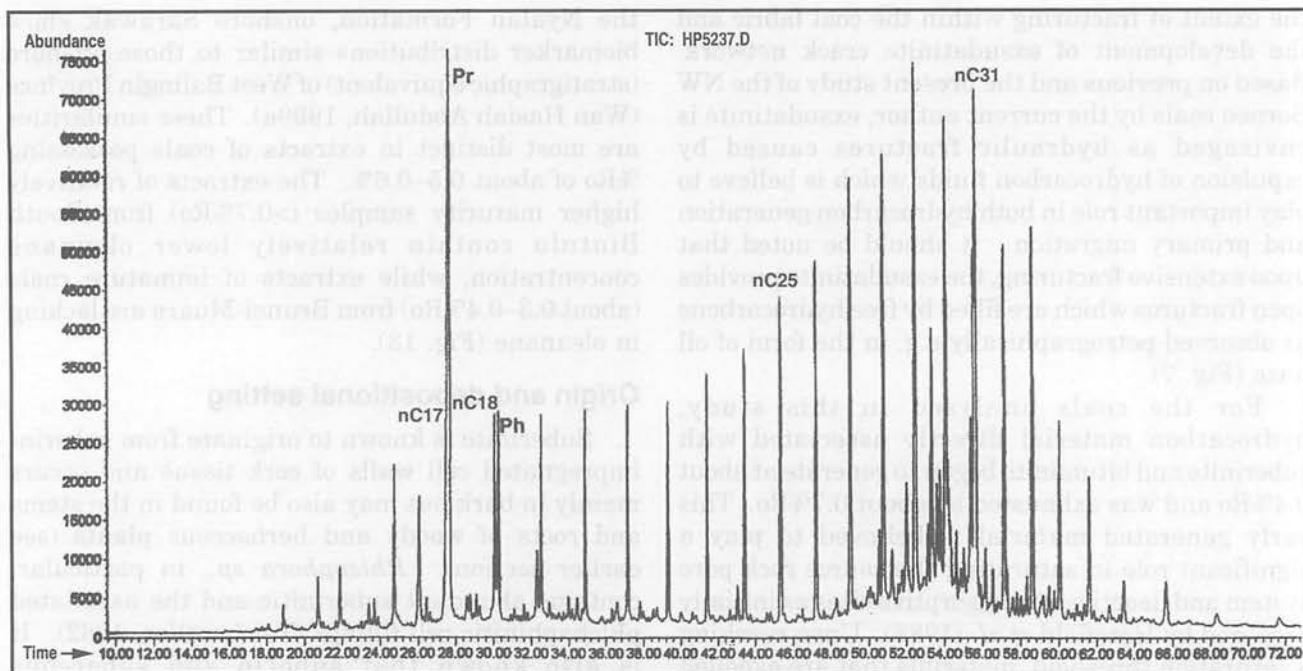


Figure 11. Gas chromatogram of saturate hydrocarbons of a Bintulu coal.

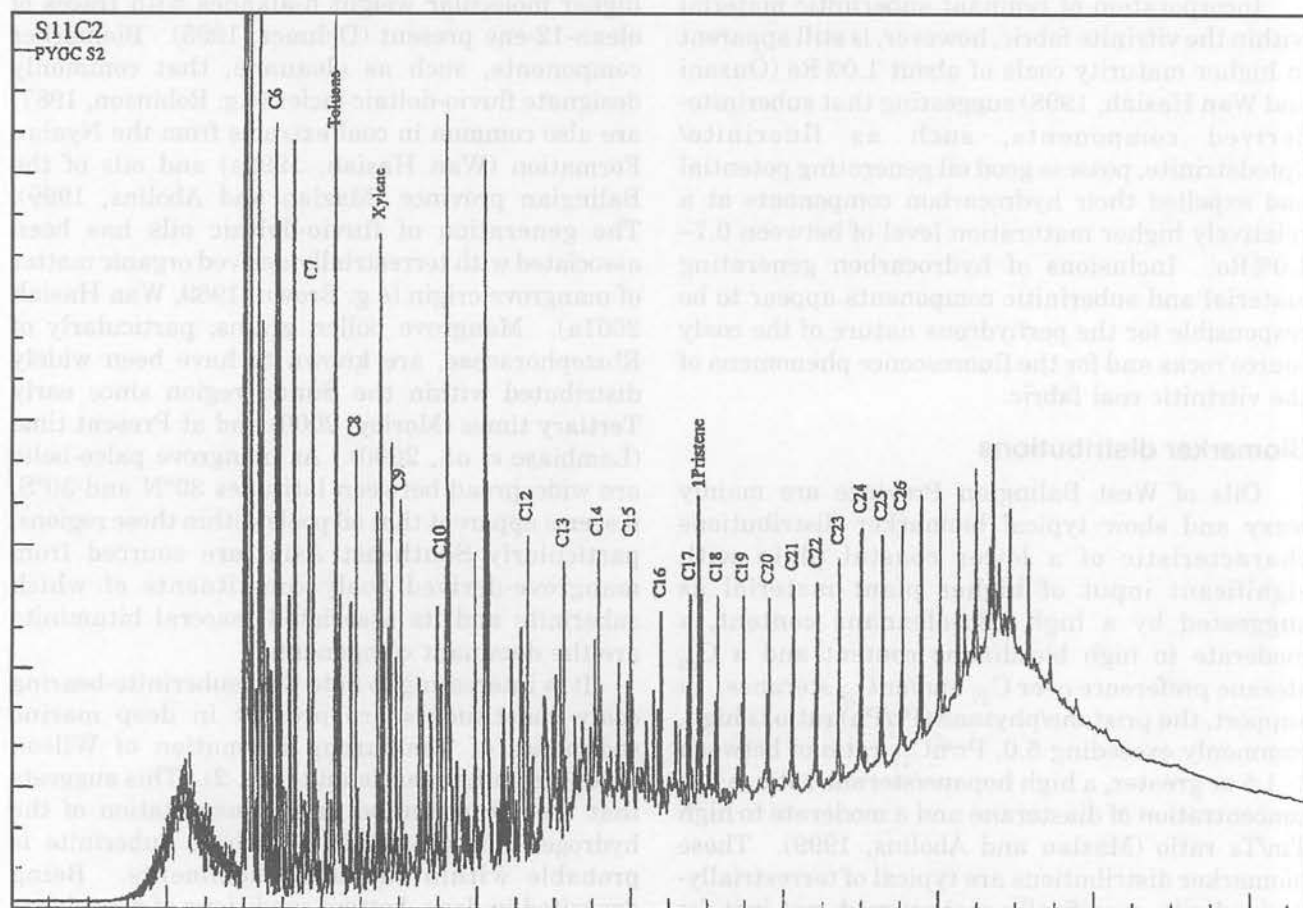


Figure 12. Py-GC (S2) pyrogram of a Brunei-Muara coal.

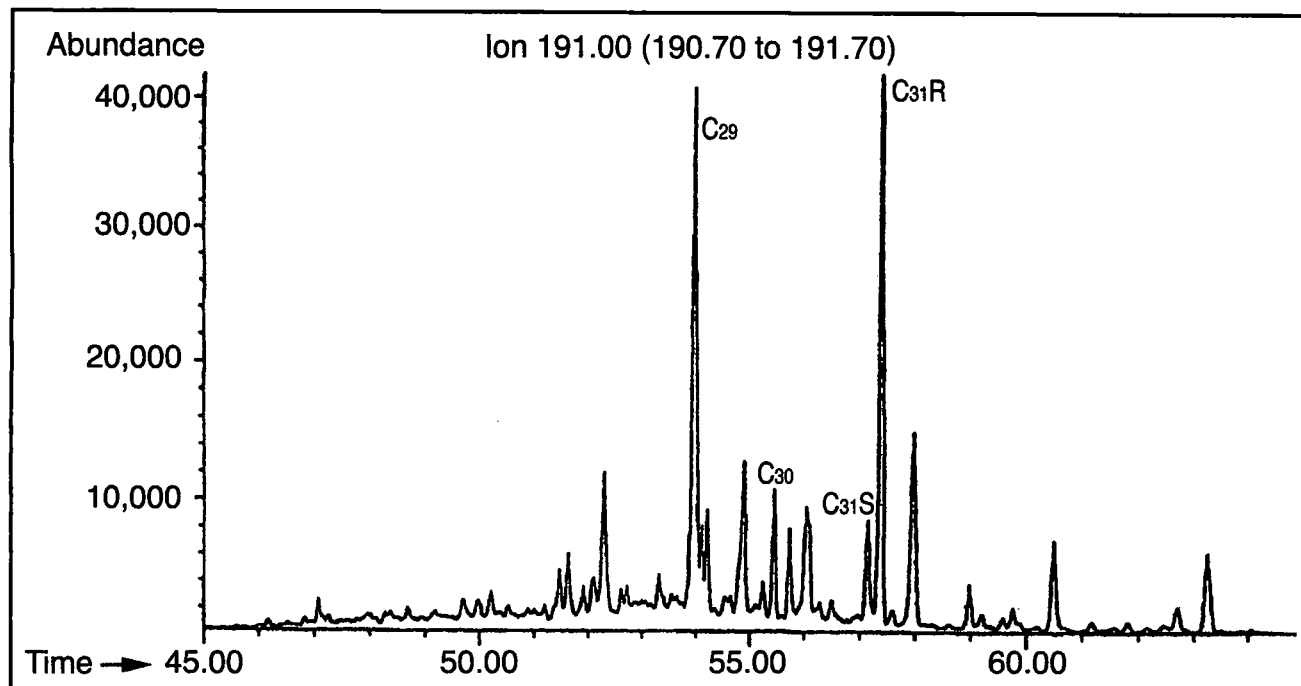


Figure 13. The m/z 191 mass fragmentogram of a Brunei-Muara coal.

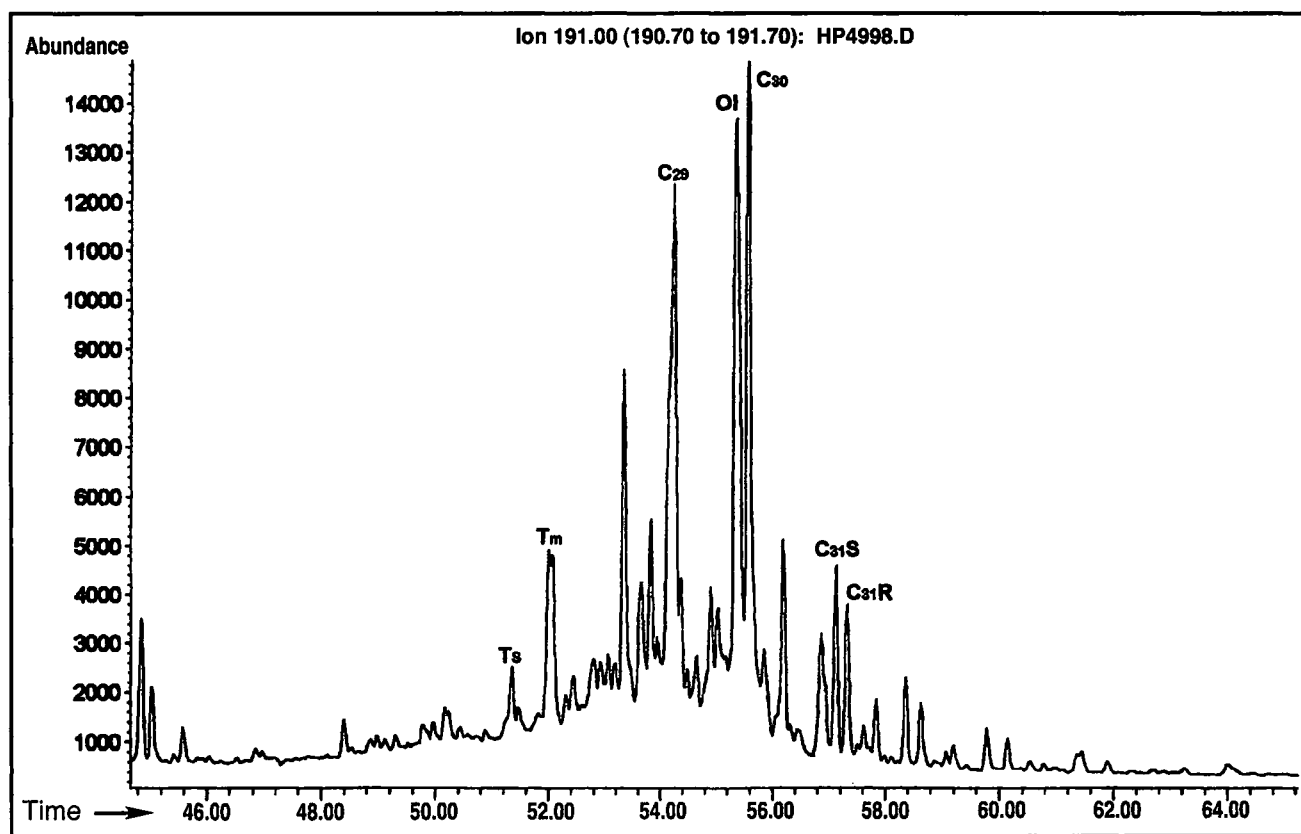


Figure 14. The m/z 191 mass fragmentogram of a carbonaceous sediment from Labuan.

their oil generation potential, possibly via bacterial reworking.

Thus, sub-environments such as mangrove swamps and lagoons within paralic and/or shallow marine settings, and some turbidite associated deep marine environments, appear to be the most likely depositional settings for these oil-prone coaly constituents.

CONCLUSIONS

Suberinite, in association with other liptinites, such as bituminite, exsudatinitite, fluorinite, liptodetrinite, phlobaphinite and weakly fluorescing vitrinite, were identified as the most oil-prone macerals of the NW Borneo coals. Bituminite is recognised here as a secondary maceral (an intermediate stage) which represents a transformation product from the precursor maceral suberinite during hydrocarbon generation. The generated or expelled hydrocarbon materials identified are mostly in the form of exsudatinitite, oil haze and oil globules.

The oil-prone macerals most likely originated from bark components and root tissues of mangrove plants such as *Rhizophoraceae* sp. and/or other suberin-bearing plant species in which suberinitic and phlobaphinitic constituents are common. Petrographic and geochemical data obtained on the samples investigated here seem to support the coaly material as mangrove-derived constituents. The biomarker distributions of the early mature coal extracts correlate well with oils of the offshore Balingian Province of Sarawak. The most distinct biomarker being oleanane, whilst the generally waxy appearance of the saturated hydrocarbon components typify these terrestrially-derived hydrocarbons. These indicate that suberinite and its associated macerals act as an important source for oils within the Balingian and Baram/Champion Delta provinces of the NW Borneo region.

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