Generation of gas and oil from coal and other terrestrial organic matter*

JOHN M. HUNT

Department of Chemistry, Woods Hole Oceanographic Institution, Woods Hole, MA 02543, U.S.A.

Abstract—Late Carboniferous—Permian sedimentary rocks contain the world's largest coal reserves which have generated and released gas but little if any oil. In comparison, organic matter deposited in lacustrine, fluvial, deltaic or brackish water sediments has sourced large quantities of waxy oils in countries such as China, Australia, the U.S.A., Argentina, Venezuela and Indonesia plus gas and condensate in deltas worldwide. Why is more oil coming from land derived organic matter disseminated in sediments than from the Carboniferous—Permian coal beds which also are land deposits? The answer appears to be in the hydrogen content and the ease of migration. When organic matter and coals have high hydrogen contents relative to carbon they can form oil. When the hydrogen is low, mainly gas is formed. Most of the world's coal is low in hydrogen whereas much of the terrestrial organic matter disseminated in sediments is higher in hydrogen so it forms more oil. Also, both gas and oil are adsorbed on coal more strongly than on the disseminated kerogen of sedimentary rocks. This makes migration of oil out of the coal more difficult compared to migration from conventional shale source rocks.

INTRODUCTION

In 1968, Hedberg wrote his classic paper on the significance of high wax oils with respect to the genesis of petroleum. Hedberg reviewed 40 regional stratigraphic sequences throughout the world which contained such oils and came up with the following conclusions:

- High wax oils are found predominantly in shalesandstone lithologies deposited under nonmarine or brackish water conditions.
- (2) Most of the sequences commonly contain coal beds, oil shales or other sediments with high organic matter contents.
- (3) The deposits are either continental, paralic, or nearshore marine environments such as lakes, bays, gulfs and deltas ranging in age from Devonian to Pliocene inclusive.
- (4) The oils produced in these sequences are high wax, low sulfur in contrast to the low wax, high sulfur crudes formed in marine environments.

Hedberg's paper was written at a time when petroleum geologists had a strong bias in favor of marine source beds. Geologists would seek a marine source whenever oil was discovered in non-marine beds even though such a source would be a long distance from the reservoir in both time and space. Since then it has been recognized that lacustrine and fluvial sediments have sourced large quantities of waxy oils in basins around the world and many of these terrestrial sediments include interbedded shale and coal sources.

Nevertheless, some bias still exists in the belief that shales are oil sources whereas coals are considered primarily gas sources.

This bias is understandable when the ultimate reserves of coal and conventional oil are compared through time. The largest reserves of coal were deposited in the late Carboniferous and Permian with lesser amounts in the Jurassic through the Tertiary (Fig. 1). In contrast, the largest reserves of oil are found in the Jurassic through the Early Tertiary (Bois et al., 1982). Ulmishek and Klemme (1990) have estimated that about 50% of the world's oil formed from Late Jurassic and Cretaceous source rocks.

Figure 1 includes both marine and non-marine sourced oils which were not separated by Bois et al. (1982). If only non-marine oils were considered, a somewhat better correlation between coal and oil might be expected. However, 75% of Hedberg's 40 world-wide high wax oil reservoirs are in the Cretaceous and Tertiary reservoirs while only 5% are in reservoirs of the coal-bearing Carboniferous and Permian periods. Consequently, other factors are related to the oil source potential of coal besides the marine vs non-marine conditions.

THE IMPORTANCE OF HYDROGEN

A limiting factor controlling the generation of oil and gas in the subsurface is the availability of hydrogen. There is no source of hydrogen other than sedimentary organic matter (OM) within the temperature range of catagenesis (about 50–200°C). The hydrogen content of OM exerts a major control on its physical properties. As the hydrogen content increases, OM goes from solid to liquid to gas. The reflectance of all coal macerals decreases as the

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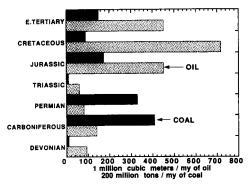


Fig. 1. Ultimate reserves of coal and oil through time.

hydrogen content increases. The pyrolysis of high hydrogen OM gives a low $T_{\rm max}$ and vice versa. Under the microscope high hydrogen OM appears lighter in transmitted light than low hydrogen OM at the same maturation level.

Table 1 shows that the carbon content of gas, oil and coal is about the same, around 80%. But there is almost three times as much hydrogen in oil and 5 times as much in gas compared to a typical humic coal. It follows that the higher the hydrogen content of coal, the greater the ability of coal to generate oil and gas.

Table 2 shows the hydrogen to carbon atomic ratio of the three major kerogen and coal types. The coal types, which are described below, have the same range of hydrogen content as the kerogens. Kerogen and coal have about the same carbon content at the beginning of catagenesis so the difference in ratios represent a difference in hydrogen content. Theoretically a boghead coal should be able to generate as much oil as the Green River oil shale, the standard Type I kerogen. Both plot in the same position on a Van Krevelen HC/OC diagram when immature (see Hunt, 1979, p. 275).

An indirect measure of hydrogen content is the hydrogen index determined by pyrolysis. In Fig. 2, petrographically identified liptinite, is plotted against the hydrogen index for 70 kerogens and coals from around the world (Mukhopadhyay et al., 1985a). There is considerable scatter in individual values in the Mukhopadhyay et al. paper but the general trend is clear. As the liptinite content of the OM increases so does the hydrogen content and the potential to generate oil as indicated by the hydrogen index.

The effect of the increasing hydrogen content on oil generation from coal has been observed in the laboratory. Lewan (1990) pyrolyzed 13 coal samples under hydrous conditions and obtained oil yields hydrogen indexing HC/gTOC

Table 1. Elemental composition of fossil fuels

	(W1%)				
	Gas	Oil	Coal		
Carbon	76	84.5	81		
Hydrogen	24	13	5		
Sulfur	0	1.5	3		
Nitrogen	0	0.5	1		
Oxygen	0	0.5	10		

Table 2. Approximate hydrogen to carbon atomic ratios of major coal and kerogen types

			<u> </u>
Kerogens	Ratio	Coals	Ratio
Type I	1.45	Boghead	1.5
Type II	1.25	Cannel	1.2
Type III	0.8	Humic	0.8

All samples are at the beginning of catagenesis.

ranging from 5 to 16 wt% on a mineral-free coal basis. The yield of high wax oils increased with an increase in the hydrogen content of the coal. Hydrogen content was the only measured factor showing a good correlation with the oil yield. Liptinite contents were not determined.

THE DIFFERENT KINDS OF COAL

Coals were originally classified as humic and sapropelic by Potonie in 1908. Humic coals which constitute over 80% of the world's coal are formed from plant cell and wall material deposited under oxic to sub-oxic conditions. Sapropelic coals, which include boghead and cannel coals, are formed from spores, pollen and algae deposited under sub-oxic to anoxic conditions (Hunt, 1979, pp. 273–279). Some boghead coals are composed almost entirely of algal colonies (Stach et al., 1982, pp. 173–177).

The major maceral groups of coal are liptinite, vitrinite and inertinite. Figure 3 shows these groups, plus individual macerals and plus wax, plotted on a Van Krevelen, H/C-O/C diagram.

Unfortunately, current books on coal petrography such as Stach et al. (1982) tend to include wax and resins together under the maceral resinite. This is probably because resins can bodentified petrographically, but waxes are invisible coatings on spores, pollen and cuticles and are unrecognized within the matrix of coal. However, waxes cannot be grouped with resins as they are entirely different in origin and composition. Waxes are formed as esters of long straight chain alcohols and fatty acids. They also contain long straight chain hydrocarbons. In contrast, resins are formed from terpenes so they contain multiple ring structures, cycloalkenes and aromatics. Waxes have H/C atomic ratios close to 2 whereas the ratios are about 1.5 for resins in the immature state.

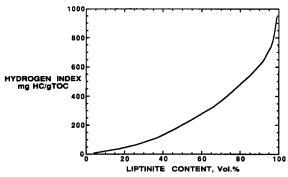


Fig. 2. Trend of increasing hydrogen index with liptinite content of kerogens and coals.

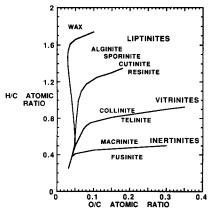


Fig. 3. Van Krevelen diagram of coal macerals and wax.

Waxes can generate much more oil than resins because of their higher hydrogen content. Van Krevelen recognized these differences by calling wax "cerinite" and classifying it under liptinites. However since this is not recognized by the International Committee for Coal Petrology, the term wax will be used in this paper.

All of the liptinites and wax are high in hydrogen in the immature stage. Wax has more hydrogen than all of the macerals (Van Krevelen, 1961, p. 117). Algae, pollen, spores and cuticles contribute wax to boghead coals and lacustrine oil shales. True cannel coals contain more sporinite than alginite. Resinite and cutinite are found in most coals and both contain up to 11% hydrogen which is more than double the hydrogen content of a typical humic coal (Table 1).

The lines of Fig. 3 represent the maturation pathways of coal macerals as originally determined by Van Krevelen (1961). Later Tissot et al., (1974) developed maturation tracks for kerogens which have only minor differences compared to the original tracks of Van Krevelen. For example, the boghead coals shown in Fig. 4 have an H/C-O/C position almost identical to Green River oil shale and follow a similar maturation pathway. Although sapropelic coals have the potential to be major oil generators

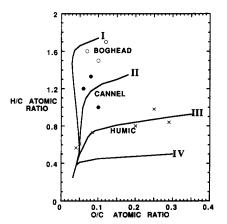


Fig. 4. Van Krevelen diagram of coals and kerogen Types I, II, III and IV: ○, boghead; ●, cannel; ×, humic.

they represent < 10% of the world's coal. Over 80% of all coals are humic and over 70% of most humic coals consist of the maceral vitrinite.

In the intermontane basins of Thailand there are oil shales up to 1 m thick interbedded with subbituminous coals. The oil shales formed when peat swamps close to a steep basin margin were flooded by shallow lakes permitting algae to dominate the organic facies. During the more oxic, regressive periods, peat swamps were left behind to form humic coals. The formation of oil shale and coal alternates with transgressive and regressive cycles. (Gibling et al., 1985).

The problem with any classification system such as that for coal macerals and coal types is that the pure individual components are rare in nature. Bogheadcannel coals are more common than either individual type. Likewise humic coals contain appreciable quantities of liptinitic materials as solid fillings. Humic coals which appear to be made up primarily of vitrinite and inertinite under ordinary reflected light often show fluorescence of vitrinites and even some inertinites indicating lipoid OM is within these macerals. Although the maturation pathways in Figs 3 and 4 were defined with well characterized kerogens and coal macerals, most of the kerogens and coals in nature fall in between the lines. The question is not whether humic, cannel or boghead coals can generate oil but rather what is the minimum liptinite, or fluorescent OM content required to make any coal a source of oil?

COAL AS A SOURCE OF OIL AND GAS

Coal has long been recognized as a source of gas, primarily methane and carbon dioxide but its importance as a generator of economic accumulations of oil is difficult to prove as coals are often interbedded with shales which may be the source beds. There are some basins in the world, however, where the evidence is pretty strong that coal is contributing to accumulations of liquid petroleum. In Fig. 5, the percentage of exinite (sporonite and cutinite) and resinite in coals is plotted against the H/C atomic

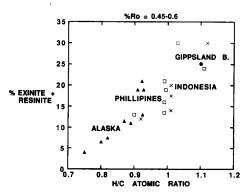


Fig. 5. Volume % exinite plus resinite vs H/C atomic ratio of coals: ♠, Alaska; □, Phillipines; ×, Indonesia; ♠, Gippsland Basin (Jones, 1987).

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ratio of the coals which is a rough indicator of their oil generating capacity (Jones, 1987). When OM has an H/C ratio larger than about 0.9, it usually indicates some liquid generating capability (Hunt, 1979, p. 275). Figure 5 suggests that immature coals with more than about 15–20% exinite plus resinite are capable of generating oil. This would include the Gippsland Basin coals of Australia and some of the coals of Indonesia and the Phillipines.

In the Mahakam Delta of Indonesia, Huc et al. (1986) found no significant difference in the maturation pathways of coals and kerogens isolated from shales interbedded with the coals. In some wells the coals were richer in hydrogen than the kerogens but all tended to follow the Type III kerogen track in Fig. 4 starting at an H/C ratio around 0.9. The yield of hydrocarbons from the coals was actually greater than from the shales based on the hydrogen index determined by Rock-Eval pyrolysis. Huc et al. concluded that there is no reason to favor shales over coals as potential source rocks in the Mahakam Delta based on their hydrocarbon potential.

Huc et al. did note that the Miocene Kerbau coals at Mahakam did not generate an extractable bitumen intermediate on pyrolysis whereas the shales did produce bitumen which was later cracked to light hydrocarbons. Also, shales in the Paris and Douala Basins produced bitumen intermediates but the coals did not. Sterane and triterpane biomarker patterns of oil extracts from thin coal seams and clays at Mahakam were found to be identical to the patterns for oils in reservoirs above (Schoell et al., 1983).

Thompson et al. (1985) examined hydrogen rich coals in three Indonesian basins, Kutai, northwest Java and Sunda. Coal samples from these basins gave hydrogen index values in the range of 250-450 mg HC/g TOC (total organic carbon). OM with HI values above 200 are generally considered capable of generating some liquid hydrocarbons. For example, the Mowry Shale, a well known Cretaceous source rock of the Rocky Mountains, U.S.A., has HI values in the 200-400 range (Jones, 1987). Microscopic analysis of the Indonesian coals indicated that they contain desmocollinite, a low reflecting, hydrogenrich vitrinite. Layers of liptinitic material were interspersed in vitrinite along with impregnations of bitumen and other fluorescent materials. The coals were estimated to contain 15-65% liptinite (Thompson et al., 1985).

The crude oils of these Indonesian basins have the characteristic odd—even predominance in the C_{27} — C_{33} range of n-alkanes along with a pristane to phytane ratio >5 which is typical of an oxic depositional environment. Mass fragmentograms of extracts from the coals showed oleanane and C_{30} — C_{31} hopane patterns similar to those of the crude oils believed to be generated by the coals. Oleanane comes from the flowering plants that were widespread in Indonesia during Tertiary times. The authors concluded that a process of liptinite enrichment that is currently

occurring in Indonesian deltas was responsible for the formation of coals as oil source rocks (Thompson et al., 1985).

Vitrinite itself can vary in hydrogen content beyond the 5% level of most humic coals. For example, Newman and Newman (1982) noted a difference in vitrinite reflectance of 0.28% between two New Zealand coal seams separated by only 100 m. The lower reflecting coal was formed in swamps with high bacterial activity and little drainage which produced a hydrogen-rich vitrinite. Such vitrinites fluoresce in reflected blue light from dull yellow to orange-brown in color indicating that they have some oil generating capability. The higher reflecting coal was from a well-drained swamp facies and did not exhibit fluorescence. Consequently, humic coals which are mostly vitrinite may generate oil from their hydrogen-rich liptinite cell fillings and laminations or from an increase in hydrogen of the vitrinite itself due to more anoxic conditions at burial.

The Gippsland Basin of Southeastern Australia had initial reserves of about 3 billion barrels of oil along with 8 trillion cubic feet of gas and 800 million barrels of condensate. The oils are high wax, low sulfur with C₂₉ steranes dominant and pristane/phytane ratios usually above 5 (Shanmugam, 1985). These are all characteristics of oils originating from OM deposited with terrigenous sediments. Sesqui- and diterpanes along with other land derived biomarkers also have been identified in the oils (Philip and Gilbert, 1986).

The source of these oils is believed to be in the Lower Tertiary Latrobe coal measures which contain wax structures plus resinite, cutinite, sporinite and suberinite. Smith and Cook (1984) divided the coal and carbonaceous shales of the Latrobe group into the Latrobe Valley facies, and Upper and Lower Eastern View facies. The latter two, which are the most likely source beds, have liptinite contents ranging up to 25% with an average around 15%. The vitrinite averages about 80% and inertinite 5%. Thick coal seams grade into carbonaceous shales and sands. The shales contain disseminated coaly matter of the same composition as the coals. Although the vitrinite content is high much of it consists of low reflecting, hydrogen-rich perhydrous vitrinite that fluoresces under reflected blue light. Consequently, much of the oil may be coming from both the liptinite and the perhydrous vitrinite (Smith and Cook, 1984).

Shanmugam (1985) reported hydrogen index values ranging from 201 to 312 for coal samples from the Halibut Field offshore and 391–422 for Latrobe coal seams onshore. These values are within the oil generating range of known shale source rocks as previously mentioned.

Although the Latrobe Valley group contains some of the world's thickest coals seams (up to 165 m according to Shanmugam) the proportion of coal in the Latrobe group decreases from about 50% onshore to about 5% offshore. The dispersed OM in

shales and sands between the coal seams offshore is nearly all Type III with maceral compositions similar to the coals. Shanmugam (1985) also carried out hydrous pyrolysis of coals and resin bodies from the Latrobe group. He concluded that the more paraffinic oils in the Gippsland Basin were derived from the wax in coals while the naphthenic oils were formed largely from the resin bodies.

The Permian-Triassic sediments of the Cooper-Eromanga Basin in South Australia comprise several cycles of fluvial sandstones, coal measures and lacustrine shales. All hydrocarbons in the basin are believed to have originated from these terrigenous sediments. There are about 100 gas fields and 10 oil fields in the basin with recoverable reserves of 5 trillion cubic feet of gas and 300 million barrels of condensate and oil. The Permian coal measures have long been considered the major source of gas but not necessarily the source of oil (Vincent et al., 1985).

Dispersed OM of the shales and macerals of the coals are predominantly vitrinite and inertinite with liptinite (mainly sporinite and cutinite) < 10%. The Permian Toolachee Formation has hydrogen indices as high as 320 suggesting some capability for liquid generation. However, the oils are found in the overlying Eromanga Basin where there are much better source rocks, namely, the Jurassic Birkhead and Basal Jurassic Formations with dispersed liptinite giving HI values up to 450. These formations contain sporinite, cutinite, resinite and suberinite all of which have oil source potential (Vincent et al., 1985). All crude oils analyzed but one are paraffinic with low sulfur contents, and the biomarker patterns such as a high C₂₉ content are characteristic of oils derived from terrigenous source rocks.

The Sand Wash Basin of Northwest Colorado produces oil in the White Dome and Powder Wash Fields which appears to be derived from thick coal beds in the Upper Cretaceous and Tertiary (J. Clayton, personal communication). But more analyses are needed to verify the correlation. Small amounts of oil also have been obtained during coal bed production of methane in areas such as the San Juan Basin.

TERRESTRIAL KEROGEN AS A SOURCE OF OIL AND GAS

Kerogen disseminated in lacustrine and fluvial shales has become well established as a source of both waxy oil and gas in the years since Hedberg's paper. Source rocks vary from the Type III kerogens found in deltas such as Mississippi, Mahakam, Niger and Mackenzie to Type I and II kerogens found in the oil shales deposited by anoxic lakes. The lacustrine reservoirs in the Songliao and Bohai basins of China which today produce over 80% of China's oil contain high wax oils which were unknown at the time of Hedberg's paper.

A comprehensive review of the petroleum geochemistry and depositional environment of lacustrine source rocks has been published by Powell (1986). A key point of Powell's paper is that there is a wide variation in the TOC from <1 to >20%, and in kerogen types from I to III and in the source of organic matter (land plant, algal or bacterial) in lacustrine sequences. Most crude oils are paraffinic and waxy due to OM sources such as pollen, alginite, cutinite and sporinite. More naphthenic oils tend to come from resinite as previously mentioned.

There is a misconception in the literature that lacustrine source beds contain primarily Type I kerogen. This is partly based on the fact that the original Type I kerogen standard defined by Tissot et al. (1974) was the Green River oil shale. But lacustrine sediments are notoriously variable in their composition so while one Green River sample may be Type I another will be Type II or III. Figure 6 shows an HI/OI plot for the kerogens of lacustrine Green River shales and the laterally equivalent alluvial Wasatch rocks of the Uinta Basin, Utah (Anders and Gerrild, 1984). The large variation in kerogen type is due to the fact that the Green River formation is a mixture of open lacustrine, marginal lacustrian and alluvial facies all of which contribute different kerogen type mixtures. The deeper open lacustrine facies samples are concentrated at the top of Fig. 6 around the Type I line and between I and II. More mature samples are further down the graph between kerogen Types I and III. Four samples of the marginal lacustrine facies are between Types I and II while the rest, including several immature samples, are between II and III. The alluvial samples are all below an HI of 200 suggesting no oil generating capability.

In the Songlia Basin of China, Yang et al. (1985) concluded from pyrolysis analysis of hundreds of samples that Type II and its mixtures were the dominant kerogens, the Type I being rather rare. This appears to be the case in other oil producing lacustrine sequences.

Powell (1986) noted similarities between the Songlia and Uinta Basins and other areas in that the Type I kerogen was restricted to part of the deepest

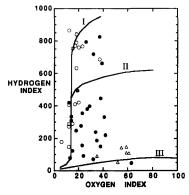


Fig. 6. Hydrogen Index/Oxygen Index plot for open lacustrian (○), marginal lacustrian (●) and alluvial facies (△) of the Green River-Wasatch formations of the Uinta Basin,

Utah (Anders and Gerrild, 1984).

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lake facies with the Type II and III increasing from the basin's center to the edge of the lake deposits. These variations suggest that the time-temperature requirements for oil generation in the deepest lake facies will be higher than in sediments more toward the margins. Activation energies required to crack Type I kerogen are significantly higher than required for Types II and III (Hunt and Hennet, 1991).

Waxy oils and gas occur in Eocene to Miocene sediments in offshore basins west of Bombay, India. Mukhopadhyay et al. (1985b) found a high content of desmocollinite and bituminite in immature kerogens of these sediments, some containing up to 70% desmocollinite. Half of the samples studied had TOC values from 2 to 14% with the other half being below 2%. H/C ratios were high ranging from 0.9 to 1.55. Pyrolysis of the kerogens yielded waxy pyrolysates. Desmocollinite is a low reflecting, high hydrogen vitrinite maceral which does yield an oil on maturation according to Mukhopadhyay et al. (1985). The desmocollinite had a weak brown fluorescence under reflected blue light while the bituminite fluoresced reddish-orange in color. Resinite with a yellow fluorescence also was present. This is a good example of hydrogen-rich disseminated terrestrial kerogen generating a waxy oil.

In deltaic sequences such as Niger and Mahakam both the Tertiary sediments and the crude oils have been found to contain oleanane which, as previously mentioned, is considered to be a product of flowering plants. In the Mackenzie delta the OM of the sediments has the characteristics of terrestrial OM such as the presence of oleanane, a high C₂₉ sterane content and a high carbon preference index (CPI) in the wax range. In addition, bisnorlupanes were found in relatively high concentrations in the crude oils (Brooks, 1986). Analyses of the sediments showed that only one formation, the Eocene Richards, contained bisnorlupanes. Consequently, the Richards Formation is considered an important contributor to the oils of the Mackenzie basin. It contains mainly Type III kerogens typical of most deltas (Brooks, 1986).

Deltas tend to have low TOCs (<5%) and mostly Type III OM with some Type II but they still generate a lot of oil, condensate and gas. Oil shales in contrast have very high TOC's and mostly oil prone Type I and II OM yet they are rarely associated with significant oil accumulations. There are probably several reasons for this. As previously mentioned Type I kerogen requires a higher activation energy for cracking, consequently most oil shales are immature. They have not been buried long enough at high enough temperatures to generate substantial amounts of oil. Second, the oils that are generated tend to be high wax oils which need further cracking for ease of migration. Third, there is frequently little or no gas and generally little water in fractured oil shale reservoirs such as the Green River Formation in the Altamont Field of the Uinta Basin, Utah. In

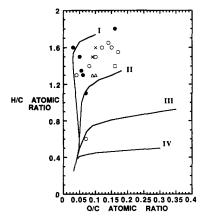


Fig. 7. Van Krevelen diagram of typical oil shale kerogens:
 ○, Lamosite; ●, torbanite; ×, tasmanites; □, kukersite and △, mixed types (Cook et al., 1981).

the absence of gas and water, there is no driving mechanism to assist the migration of a thick, waxy oil. Also, some formations like the Green River are overpressured due to the formation of a hydraulic seal which prevents the free flow of water and the migration of generated hydrocarbons (Hunt, 1990).

Figure 7 shows a Van Krevelen plot for kerogens in a series of oil shales (Cook et al., 1981). Some oil shales grade into the boghead and cannel sapropelic coals. Much of the OM of immature oil shales fluoresces under incident blue light. Nearly all of them form from algal oozes deposited under sub-oxic to anoxic conditions. Total world reserves of shale oil proved and possibly recoverable are about 3×10^{12} barrels (Cooke et al., 1981). However, recovery costs and disposal of the byproducts are such that the economic use of oil shale is still far in the future. At present only a very small percentage of the oil in the world is produced from fractured oil shale reservoirs.

ADSORPTION-MIGRATION CHARACTERISTICS OF COAL AND TERRESTRIAL KEROGEN

There is no doubt that hydrogen-rich coal and terrestrial kerogen can generate economic quantities of liquid petroleum. This has been recognized in the field and in laboratory experiments discussed in this paper. Why then do we not find more oil accumulations generated by coal beds? One problem is that most thick coal beds are humic coals relatively low in hydrogen whereas thinner beds interbedded with shales and sometimes oil shales tend to contain hydrogen-rich vitrinite or sapropelic coals rich in liptinites. But coal also has problems related to adsorption and migration phenomena which do not seem to be as critical with the disseminated kerogen or coaly particles of shales.

Charcoal is one of the most widely used adsorbents for taking bitumens and other organic contaminants out of a flowing water system. Laboratory experiments have shown that as you go from high volatile bituminous coal to anthracite, the adsorption of hydrocarbons increases (Wyman, 1984). Increasing pressure also increases the adsorption. The larger hydrocarbon molecules would be adsorbed more strongly than smaller ones since adsorption on coal would be comparable to that on a packed column in gas chromatography. Wyman (1984) slowly desorbed coal samples obtained at a depth of 2835 m in the deep Western Canada Basin. He found that after 15 days only the hydrocarbons methane, ethane and propane were desorbed. By 34 days he was also obtaining isobutane, *n*-butane and isopentane. Extraction of these coals released up to 30 mg HC/g TOC which apparently was strongly adsorbed on the coal surface.

Barker et al. (1989) studied the adsorption of hydrocarbons and asphaltenes on both wet and dry quartz, clays and carbonates. They found that there is an initial monolayer coverage which requires higher activation energies for removal than multilayers that follow. Pyrolysis of samples with adsorbed hydrocarbons causes their release between pyrolysis peaks 1 and 2 frequently overlapping peak 2, the cracking peak. This suggests that in natural systems these strongly adsorbed hydrocarbons would not come off at the end of the oil window and consequently would be cracked to gas and condensate without releasing liquid hydrocarbons.

Barker et al. (1989) also studied the adsorption of bitumens on shales. Figure 8 shows bitumen adsorption in arbitrary units plotted against TOC for a shale with a Type II kerogen. They found that above 9% TOC there was a sudden increase in adsorption efficiency. This suggests that it would be difficult for hydrocarbons to migrate out of high TOC shales or coals prior to their conversion to gas.

Another property of coals that prevents liquid hydrocarbon migration is the pore diameters. Coal pore diameters can be divided into macropores >50 nm (nanometers), mesopores 2-50 nm, micropores 0.8-2 nm and ultramicropores <0.8 nm (Walker, 1981). Coals vary widely in both pore volumes and porosities. Parkash and Chakrabartty (1986) found that 30-76% of the pore volume in 11 Alberta plains coals was in the micropore range while 19-50% was in the macropore range and the rest in

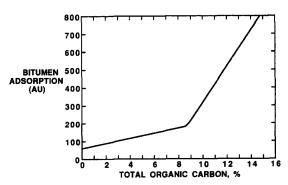


Fig. 8. Bitumen adsorption on the Excello shale. AU equals arbitrary units (Barker et al., 1989).

between. Surface areas of the coals calculated from carbon dioxide adsorption at 25°C varied between 75 and 500 m²/g. Since complex hydrocarbon ring structures are about 2 nm in diameter and small asphaltenes 5 nm, it appears that newly generated liquid bitumens would not be able to migrate out of the 30–76% micropores until the coal is fractured. Fracturing could occur when the bitumens are converted to gas.

There is other laboratory evidence that coals trap hydrocarbons as they are generated. Landais and Monthioux (1988) carried out open and closed pyrolysis of coals. The same coal samples followed different maturation pathways using open, confined and closed system pyrolysis. Comparing these results with extraction data they concluded that natural coals keep free hydrocarbons trapped in the pores of their structures.

Youtcheff et al. (1983) found that the quantity of alkanes released from coals by liquifaction in tetralin at 400°C is 6-8 times greater than the yield obtained by Soxhlet extraction. They concluded that the alkanes obtained by liquifaction had been physically trapped inside the pores of the coals and were only released when these pores were broken open by liquifaction.

In recent years oil companies have begun producing methane directly out of coal beds as well as sands associated with the coals. One observation of this new approach is that some wells produce nothing but water for the first few weeks or months. As the coal dewaters, however, the gas starts coming in faster with less and less water. This dehydration process results in desorption of hydrocarbons from coals and increased fracturing of the coals. Gas production actually increases rather than decreases with time as the pressure goes down. This is further evidence that fracturing of pores and desorption are critical factors in getting hydrocarbons out of coal. Apparently it is more difficult for liquid hydrocarbons to migrate than gases so the liquids stay in place until they are converted to gas.

CONCLUSIONS

- (1) Coals and terrestrial kerogen with H/C ratios above about 0.9 or hydrogen indices above about 200 by Rock-Eval pyrolysis, and liptinite contents above 15% have the potential to generate and release oil as well as gas. Typical examples are oil fields in Indonesia and Australia.
- (2) Terrestrial kerogen, deposited in lacustrine, fluvial, deltaic or brackish water sediments, ranges from Type I to III with major oil production coming from Type II in lacustrine environments such as the Songlia Basin of China and from Type III in deltas.
- (3) Pore size distributions and adsorption characteristics of bedded humic coals appear to prevent the release of generated bitumens. Their eventual conversion to gas and condensate appears to cause

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the fracturing of the fine pores and release of trapped hydrocarbons. Desorption of the smaller molecules formed by cracking is probably assisted by CO₂ and CH₄. Disseminated Type III kerogens and coaly particles do not seem to trap and adsorb hydrocarbons as strongly as bedded coals.

(4) More studies are needed on hydrocarbon adsorption-desorption characteristics of thick and thin coal beds and shales having a range of TOC values and H/C ratios. Also, comparisons are needed between the extent of hydrocarbon trapping in bedded coals compared to disseminated Type III kerogen.

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