

Influences of coal composition on coal seam reservoir quality: A review

by

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ABSTRACT

Economic production of natural gas from coal seams requires the satisfaction of three conditions: 1) sufficient quantities of methane were generated during coalification, 2) sufficient quantities of methane have been retained within the coal, and 3) occluded gases can desorb and migrate through the reservoir at economically acceptable rates. These three requirements are influenced by coal composition and are discussed on the basis of three criteria: grade (the proportion of mineral vs. non-mineral constituents), type (the diverse varieties of organic (non-mineral) constituents), and rank (the level of compositional maturity imparted during diagenesis).

Generation

The quantity of methane formed during coalification is dependent upon initial coal composition. Coals rich in inertinite and subhydrous vitrinite will generate less hydrocarbons than perhydrous coals. Mineral matter does not directly generate hydrocarbons, but may catalyze some types of organic maturation reactions and may also influence expulsion. Expulsion of higher hydrocarbons and oil diminishes the amount of methane formed. Hydrocarbons are generated throughout coalification, but expulsion rate appears to increase rapidly around the transition from high vol A to med. vol. bituminous rank, or the bituminization to debituminization stage of coalification (approx. 0.95% R_{vo} and 82% C), when other reservoir-related properties of coal pass through an inflection.

Retention

Gas sorption capacity of coal is directly proportional to organic matter content. Low-ash coals generally provide the most attractive production targets but bed thickness must also be considered. Laboratory studies

suggest that the maceral group vitrinite has the highest sorbate accessibilities, yet preliminary field data suggest inertinite content correlates positively with desorbed gas yields. With increasing rank, total sorbate accessibility, including H₂O, CH₄, and CO₂, initially decreases, passing through a minimum around med vol to semianthracite (85-90% C), then increases through anthracite rank. Coal hydrophobicity increases with increasing rank resulting in a net increase in methane sorption capacity with rank for water-saturated vitrinite-rich coals.

Migration

Diffusion rates appear to be somewhat faster for high ash coals than for clean vitrinite-rich coals, and are comparatively rapid for OM-rich shales. Occluded methane has been successfully produced from some thick organic matter-rich "black" shales, which exhibit reservoir behaviour similar to coals. Fracture spacing, which varies inversely with mineral matter content and is more closely spaced in vitrain bands and vitrain-rich coals, influences effective permeability and hence, desorption rates.

INTRODUCTION

Underground coal seams throughout the world contain an enormous resource of methane and other light hydrocarbons occluded within their molecular structure. With appropriate production technology, a portion of these hydrocarbons can be liberated in the form of natural gas, along with variable amounts of other entrapped species such as carbon dioxide and nitrogen. It is the goal of most commercial coal seam degasification projects to produce this gas at rates and volumes that render it economically competitive with other more "conventional" sources of natural gas. When coal degasification is

done in conjunction with underground mining, however, additional economic incentives may come via reductions in ventilation costs, reduced explosion hazard, and reduced equipment down time. A further, non-economic incentive for degasification in advance of mining is the reduced contribution of mine methane emissions to the so-called "greenhouse" gases in the atmosphere.

A decade or so of (occasionally frenzied) drilling and production of coal seam gas in North American basins has demonstrated that reservoir characteristics of coal are highly variable and heterogeneous at many different scales. Satisfactory production rates have only been encountered (for the most part) in restricted parts of just two North American basins, the northern San Juan basin (New Mexico and Colorado) and the eastern Black Warrior basin (Alabama). As efforts to produce coal seam gas are expanded into other parts of North America, Australia, and the world, it is important to gain a better understanding of the geologic and engineering factors controlling the reservoir properties of coal, especially those affecting production rates. If these factors and their degree of variability can be identified then models can be developed for better production and further exploration and development.

While much is already known concerning the influence of coal composition on the technological behaviour of coal (e.g. coking, combustion, liquefaction, and gasification), comparatively little is understood regarding reservoir properties; and whereas the properties and behavior of conventional reservoir rocks have been intensively studied for many decades, the reservoir properties of coal have only recently begun to be carefully studied. Coal analytical techniques such as proximate analysis, which have commonly been used in the past to evaluate the technological characteristics of coal are of limited value in evaluating coal as a reservoir stratum and new techniques must be developed. However, useful results can be gleaned from the presently available literature.

AN OVERVIEW OF COAL COMPOSITION

As a sedimentary rock, coal composition can be described in terms of three fundamental characteristics: 1) grade, which represents the relative proportion of organic vs. mineral constituents, 2) type, which represents the diverse varieties of organic (non-mineral) constituents, and c) rank, which represents the level of compositional maturity imparted during diagenesis, beginning with peat and continuing through brown coal (lignite), bituminous, and anthracite. Grade classification is normally based upon the weight percentage of organic matter present. Type designation is usually based on visual classification of the macroscopic banding character or the microscopic organic constituents of coal (the "macerals"). More recently, dominant organic-geochemical parameters are used to type coal. Rank can be determined chemically by fixed carbon yield (dry, mineral matter-free), or any number of other rank-dependent variables, or optically by vitrinite reflectance. However, each rank parameter is subject to inaccuracies related to differences in organic matter (OM) type. The diagram in Figure 1 is intended to provide a comprehensive scheme by which all coals and/or OM-bearing sedimentary rocks can be represented, with grade, type, and rank being depicted on three orthogonal axes.

The grade, type, and rank of a coal are reflective of the geological processes acting throughout its deposition and burial history. On a regional and mine site scale they can vary laterally within single coal beds or between stratigraphically different seams. Together, "type" and "grade" are initially established in the sedimentary environment of deposition, although both undergo subsequent modification in the burial environment. Thus, in Figure 1, the plane defined by the "type" and "grade" axes corresponds to the environment of deposition. The projection of this plane into the third dimension ("rank") corresponds to the subsurface "burial environment".

Grade

"Grade" takes account of the fact that sedimentary rocks occur across a continuum of compositions, from clean organic matter at one extreme to pure mineral matter at the

other. Although many sedimentary rocks are essentially devoid of organic matter, no coal is entirely free of inorganic components. As specified in many international definitions, the term "coal" is reserved for those rocks comprised primarily of organic constituents, with the delineation between coal and non-coal typically set at or near 50 wt-%.

The term "ash", which is commonly and erroneously equated with "mineral matter", refers to the solid residuum remaining after combustion or low temperature ashing. Ash is largely derived from mineral matter, but can be quite different compositionally. The term "ash yield" should always be used in lieu of "ash content".

While the present paper deals strictly with "coal seam" reservoirs, it should be noted that the behavior of some carbonaceous shale gas reservoirs, such as the Devonian Chattanooga-Ohio-Antrim trend in the eastern U.S., is apparently very similar to coal seam reservoirs in terms of gas storage mechanism (by occlusion in the organic matter) and gas migration (by flowing through networks of interconnected fractures). The principal differences are in the concentrations of occluded gas in the reservoir (typically much lower in shale reservoirs than in coal) and the thickness of the pay zone (typically much greater for shale reservoirs)(Lang and Oliver, 1991).

Type

"Type" takes account of the fact that coal is a heterogeneous "plum pudding" mixture of many different organic constituents, having widely varying compositions and physical properties. The purpose of type classification is to provide a systematic method of subdividing this heterogeneous mixture into a manageable number of meaningful groups or classes. OM types present in coal are classified in many different ways, depending on the scale of measurement and the analysis method employed. For example, coals and coal constituents may be described according to physical differences recognizable at hand-specimen scale (e.g. banding or "brightness" profiles), or according to microscopically discernible color and morphology (e.g. maceral or "petrographic" classification), or according to organic geochemical composition (e.g. aromatic vs. aliphatic carbon, low molecular

weight vs. high molecular weight components, lignin-derived vs. lipid-derived components, macromolecular matrix vs. molecular fraction, etc.). Coal maceral composition provides a useful guide to gross differences in elemental and molecular composition and should always be considered in coal analysis. The sorption of gases, however, takes place at a nanometer scale, so the geochemical composition and structure should also be evaluated directly.

The various "type" constituents comprising coal reflect the combined influence of: 1) the inherent variability of the biological materials initially contributing to the coal deposit (plants and micro-organisms) and 2) the degree and nature of mechanical and (bio-) chemical degradation occurring shortly after deposition. Both factors have varied significantly through geologic time and in response to differences in climatic setting and depositional regime, giving rise to coal deposits of greatly differing chemical composition and physical properties. Of particular importance in this regard is the occurrence of coals rich in inertinite constituents in parts of Australia and other parts of the world. These coals contrast markedly with the vitrinite-rich coals typical of eastern North America and western Europe. Much of the presently available data on gas production characteristics of coal is based upon these vitrinite-rich coals. Some data suggest that the influence of different coal types on the reservoir properties of coal are at least as strong as the influence of grade and rank, and should certainly always be considered in reservoir characterization.

Rank

The concept of "rank" takes account of the fact that, beginning with its deposition on the earth's surface and continuing throughout its burial history, coal and other sedimentary OM undergoes a progressive evolution in composition and structure, collectively termed "organic metamorphism" or, when referring specifically to coal, "coalification". The physical and chemical changes associated with organic metamorphism largely represent the response of sedimentary OM to elevated temperatures and pressures in the subsurface. However, oxidation and bacterial degradation may also be important processes

in the near-surface environment. Coalification reactions that are activated merely by elevated temperatures are referred to as "thermogenic", while those that are catalyzed by microbiota are referred to as "biogenic". Recent studies suggest that biogenic processes may be more important in their influence on shallow coal bed reservoirs than has been generally recognized in the past.

A variety of physical and chemical parameters may be used to provide a measure of how far the coal has advanced along the coalification pathway (Fig. 1). Although vitrinite reflectance is now the single most widely used indicator of rank, none of these parameters is universally applicable and/or free of complications relating to coal type.

Coal rank profoundly influences the coal bed reservoir in two fundamental respects: 1) The processes of coalification (both thermogenic and biogenic) are associated with the loss of water and the generation of oil and gas in the subsurface. Thus, a substantial proportion of the methane, carbon dioxide, and other occluded volatile components of coal may have been generated from the coal itself as by-products of coalification, and 2) All of the physical and chemical properties of coal undergo a substantial alteration during coalification. These changes, in turn, influence the reservoir characteristics of the coal.

Matrix vs. Molecular Constituents of Coal

Although the fundamental characteristics of coal molecular structure have been known since the 1950s (Van Krevelen, 1961), research conducted over the past two decades, particularly in regards to coal conversion technologies, has provided a considerably more detailed, and in some respects markedly different picture of coal structure. The older view depicted coal as a three-dimensional polymeric structure, comprising polycyclic aromatic centers, fringed by functional groups, and interconnected by a network of covalent bridge structures (Davidson, 1982; Given, 1984; Behar and Vandenbrouke, 1987). The process of coalification was envisioned as representing primarily a process of condensation-type polymerization reactions, by which low molecular weight substances, including CO₂, H₂O, CH₄, and other n-alkanes were generated as by-products.

The polymeric model for coal is still basically valid, but it is now generally recognized that a diverse suite of "smaller" molecules are entrapped within this three dimensional structure, ranging in size and complexity from CH₄, CO₂, and H₂O at the low end to high molecular weight aromatic tars and asphaltene at the upper end. Strong covalent bonds are much less important in binding this mass together than was previously imagined (Larsen and Wei, 1988). Instead, weaker, hydrogen bonding and van der Waals bonding are important. This model is best described as a "two-phase" system, comprising a three-dimensional, covalently bonded "macromolecular matrix", plus an entrapped "mobile phase" or "molecular fraction" (Vahrman, 1972; Davidson, 1982; Given, 1984; Given *et al.*, 1986; Derbyshire *et al.*, 1989; Herod *et al.*, 1991; Fig. 2). The smaller of these entrapped molecules are relatively free to detach from the remainder and migrate through the coal structure. Of these, CH₄ is of particular economic significance as it may be produced in the form of natural gas. The larger molecules, however, tend to remain bonded or physically entangled in the network unless acted upon by thermal or strong solvent extraction methods. Some recent researchers have gone a step further, and have questioned whether there is any significant amount of covalent bonding within the coal matrix, suggesting instead that entanglements and weak secondary bonds are sufficient to hold the mass together (Nishioka, 1992). This distinction is (arguably) of little direct relevance to coal bed reservoirs, however, since in natural settings, these larger molecules are effectively immobile.

An additional modification of the older models is the recognition that coalification, particularly at peat through high volatile A bituminous rank, proceeds more by depolymerization than by condensation-type reactions (Hatcher and Spiker, 1988; Larsen and Wei, 1988). The net result is that the residual coal is broken into progressively smaller pieces by thermolytic cleavage of covalent bonds. The question of what happens to the depolymerized fragments (and/or condensation by-products) once they form is the topic of considerable current debate. The dramatic increase in pyridine extract yields (Larsen and Wei, 1988), fluorescence characteristics (Teichmuller and Durand, 1983), Rock-Eval

pyrolysis yields (S1 plus S2; Durand and Paratte, 1983), as well as other evidence, demonstrates that a substantial proportion of the freed molecules remain physically entrapped and/or weakly bonded within the coal. On the other hand, the changing bulk elemental composition of (dry) coal over this rank interval (mostly representing a decrease in O/C ratio in vitrinite-rich coals, accompanied by slight decrease in H/C ratio) demonstrates that a portion is expelled into the surrounding strata. The quantity of material expelled and the form in which it is expelled has important implications for the role of coal as a source rock for oil and gas.

As a consequence of processes occurring during coalification, the composition and concentration of the molecular fraction of coal is not static, but changes markedly (Fig. 3). The principal change occurring at low rank (peat through subbituminous A) is a progressive loss of water. At low rank, water is the principal pore-filling fluid in coal, comprising over 75% of peats, but decreasing to 15-20% when coal enters the oil window (high volatile C/B). At the peak of the oil window, water content of some coals has dropped to less than 1%. Water loss is partly due to physical compaction, but is also due to thermal or biogenic cleavage of hydrophilic functional groups such as hydroxyl and carboxylic acid groups (Suggate, 1990). This is followed by the formation of hydrogen-rich materials of low to high molecular weight at higher diagenetic grade, a process termed "catagenesis" by Tissot and Welte (1984), or bituminization by Teichmüller (1987). The process of repolymerization, or (re-)combination of smaller molecules into larger pieces becomes the predominant process only at high rank (from medium volatile bituminous and beyond). This process is also accompanied by the generation and partial expulsion of low molecular weight by-products, particularly methane. It is during this stage of coalification that most (vitrinite-rich) coals begin to exhibit a significant loss of hydrogen, indicating expulsion of hydrogen-rich by-products, especially in the form of methane. It has been interpreted (if not indeed proven) that some Australian coals expel oil in early stages of maturation (Cook and Struckmeyer, 1986; Saxby and Shibaoka, 1986).

Although water is sometimes not viewed as a bona fide constituent of coal, inherent moisture shares the same sort of interrelationship (especially involving hydrogen bonding) with coal structure as other so-called "organic" molecular constituents. When moisture is removed, the coal structure collapses and shrinks, and all measurable physical and chemical properties change. In high rank coals, changes associated with dehydration are (apparently) reversible by resaturating the coal with moisture (Stansfield and Gilbert, 1932; Rees *et al.*, 1939) but at low rank they are partly irreversible (Selvig and Ode, 1953). This irreversibility is apparently related to collapse of the molecular structure and formation of new intermolecular bonds within the non-hydrated fraction. Indeed, moisture provides part of the binding force that holds low rank coal together. When moisture is removed, most low rank coals shrink and crack visibly, become weak and friable. This tendency makes strength testing very difficult to perform accurately.

Methane exhibits a similar relationship to coal structure as water, although water is typically present in much higher molar concentrations. In fact (as will be discussed subsequently), water and methane "compete" with one another for accessibility within the coal structure. Similarly, it will be demonstrated that the presence of higher molecular weight hydrocarbons also effectively diminishes the coal's capacity to sorb methane and water. Thus, to be fully understood, all the molecular components should be regarded as part of a complex system.

Estimates of the weight percentage of molecular fraction present in coal are variable and somewhat ambiguous, depending on the analytical method used, owing largely to the inherent difficulty in distinguishing molecular versus matrix constituents (see discussion in Given *et al.*, 1986 and Derbyshire *et al.*, 1989). Moisture contents (which are less ambiguous) decrease dramatically from over 75% in peat to as little as 1% (or less) in high volatile A bituminous coals. The percentage of organic constituents removable by solvent extraction is more uncertain, however, and may vary widely for any given coal depending upon the solvent system used. Pyridine extract yields, which are believed by some coal chemists to provide an accurate

estimate of the entrapped molecular fraction, reach a maximum of around 25-35% for some high volatile A bituminous coals, then decrease once again at higher ranks. Coals exhibiting the highest pyridine yields correspond to a rank range equivalent to around 0.95-1.0% vitrinite reflectance (upper high volatile A bituminous). Beyond this peak, pyridine yields drop to near zero at anthracite rank, but the accessibility of small molecules increases once again, indicating a re-opening of the molecular structure.

Physical Structure of Coal

Porosity has a relatively straightforward meaning in conventional reservoir rocks, where it refers to the fraction of the bulk volume not occupied by mineral grains. Porosity is more ambiguous when applied to coal, where it constitutes a more-or-less integral part of the organic structure. In most previous investigations (e.g. Gan *et al.*, 1972; Mahajan and Walker, 1978; Mahajan, 1989) the term porosity in coal has been used in reference to the proportion of the bulk volume that can be occupied by relatively small, mobile molecules such as He or H₂O; but if the volume occupied by all of the molecular constituents is also included, then total porosity is much greater.

Coal porosity may be subdivided into two categories, macroporosity and microporosity, although it is difficult to make a clear operational distinction between the two. Macroporosity refers, in general, to openings that are too large to be considered part of the molecular structure, including cracks, cleat, fissures, vacant cell lumens in fusinite, etc. Microporosity (i.e. interstices of around 0.5 nm in dimension, is effectively a manifestation of the relatively open, poorly consolidated molecular structure of coal, which provides an intra- and intermolecular "free volume" that may either be vacant or filled by small molecules such as He, H₂O, CO₂, and CH₄. Owing to intermolecular forces acting at this scale (van der Waals forces and hydrogen bonding), gases such as methane, which penetrate into such small interstices exist in a concentrated "near liquid-like" form (Metcalfe *et al.*, 1991)

Porosity is not a fixed property of coal, but varies for different molecular probes, all of which (with the arguable exception of he-

lium) sorb to some degree on "internal surfaces" in the coal structure. Coal swells measurably during sorbate penetration, even for very weak solvents such as CO₂ (Reucroft and Patel, 1983; Reucroft and Patel, 1986), and CH₄ (Briggs and Sinha, 1933; Harpalani and Schraufnagel, 1991). Thus, sorbate molecules are not merely occupying vacancies within the coal structure, but are interacting with and causing a physical rearrangement of the coal. Moreover, the molecular sieve properties of coal (Van Krevelen, 1961) and evidence that diffusion of gases in coal is an activated process (Nandi and Walker, 1975) indicate that regions within the coal structure accessible to gaseous sorbates are typically on the order of 0.5-1 nm in dimension. The physical significance of porosity and surface area is highly questionable at this scale (Spencer and Bond, 1966). In light of these issues, Franklin (1949) and Fuller (1981) have suggested use of the term accessibility rather than porosity to describe the ambiguous relationship between transient molecular species and the more stable substrate.

It has long been established that the accessibility of coals to gaseous sorbates generally decreases with increasing carbon content (for typical vitrinite-rich coals), passing through a broad minimum around 85-90 wt-% C, and increasing once again at higher ranks (Fig. 4). This relationship has been demonstrated for N₂, Kr, CO₂, Xe, He, H₂O, hexane, methanol, and benzene (Franklin, 1949; Walker and Kini, 1965; Thomas and Damberger, 1976; Nelson *et al.*, 1980). (This relationship holds for methane also, as discussed subsequently. Comparative studies show that low pressure CO₂ surface areas provide a good guide to high pressure methane sorption capacity: Williams and Smith, 1981; Lamberson and Bustin, 1992).

The loss of so-called internal "surface area" in the oil window has been interpreted as representing the progressive "plugging" of the coal microporosity by secondary hydrocarbons generated during maturation (Thomas and Damberger, 1976). The increase in internal surface area at higher rank represents a process of thermal "deplugging" of the occluded hydrocarbons (Thomas and Damberger, 1976), resulting in the generation of "secondary porosity" (Levine and Tang,

1989). The plugging effect need not be envisioned in such graphic terms. Alternatively, it might be envisioned that the depolymerized mass of organic molecules remaining at the peak of the oil window, is held together by a higher concentration of hydrogen bonds, allowing fewer accessible sites for gas sorption.

Problems Inherent in Compositional Analysis

Compositional analysis of coal is extremely difficult—a fact which cannot be overemphasized. Whereas it can be a very simple matter to collect a sample for analysis, it is much more difficult to procure a sample that will yield useful, significant, reproducible results (see Klein and Wellek, 1989). Contributing to these difficulties are coal's: 1) non-crystalline, polymeric, macromolecular nature, 2) low atomic number building blocks (mostly C, H, O, N, & S), 3) heterogeneous nature, and 4) volatility. While it is outside the scope of the present paper to discuss coal analysis problems in detail, a few points should be stressed, particularly since interpretation of reservoir behavior is dependent upon the reliability of the compositional data.

Two problems inherent in coal analysis are non-representative sampling and compositional alteration of the sample. The potential for non-representative sampling depends upon the circumstances, with some seams being highly uniform and others more variable. Analysis work in North America over the past decade has demonstrated the high degree of variability, even over very small distances, in reservoir properties. Care must be taken, therefore, to control for such variability.

The problem of alteration is a natural consequence of the sensitivity of OM (especially at low rank) to changes in the ambient conditions of temperature, pressure, and chemical environment. A coal seam which has existed for tens of millions of years in an oxygen-deficient, water-saturated environment may undergo rapid oxidation and desiccation when exposed to atmospheric conditions (Berkowitz, 1989). These changes may or may not be reversible, and may or may not significantly influence the results of analyses, depending on the particular coal. Problems such as these can be circumvented or minimized, but only with careful thought given to the proce-

dures used for sample procurement and handling. Owing to these difficulties, care must be taken in the interpretation of previously-published data as to whether sampling and analysis was done satisfactorily. Unfortunately, there are a great deal of inaccurate, imprecise, or ambiguous data published in the coal literature.

An additional problem in interpretation of analytical results relates to the statistical significance of observed relationships. In natural systems, many factors combine to influence the reservoir characteristics of coal. Simple interrelationships between two compositional variables (e.g. methane yield and vitrinite content) may be overwhelmed or obscured by the more pronounced influence of other variables (e.g. rank, depth, ash yield). Consequently, a multivariate statistical approach is commonly needed to elucidate specific interrelationships; but only rarely are the necessary financial and human resources devoted toward this end. Commonly, coal composition is conferred only cursory consideration in reservoir evaluation. For this reason, published field and experimental data typically do not permit specific interrelationships to be properly evaluated.

INFLUENCE OF COAL COMPOSITION ON RESERVOIR CHARACTERISTICS

Viewed in general terms, four conditions must be met to enable commercial production of coal bed natural gas: 1) Sufficient quantities of gas must have been generated during organic maturation, 2) Sufficient quantities of gas must have been retained in the coal bed, 3) The gas must be able to migrate through the bed at an acceptable rate, and 4) The reservoir must be large enough to justify the cost of development. Of these, the first three are directly influenced by coal composition.

The following sections examine the influence of coal composition, as reflected by grade, type, and rank, on the three requirements for economic production, i.e. generation, retention, and migration. Interactions are evaluated both from the standpoint of theoretical relationships and currently available empirical data.

Hydrocarbon Generation

Hydrocarbons formed as by-products of coalification are partially retained within the macromolecular matrix and partly expelled. It is difficult in practice, therefore, to isolate the questions of generation and retention, owing to their interconnectedness (Juntgen and Karweil, 1966). Similarly, the influences of type and rank cannot be considered independently, as various coal constituents behave quite differently during maturation in terms of the quantity and type of by-products formed and released.

Previous estimates of hydrocarbon generation from coal (e.g. Juntgen and Karweil, 1966) have generally assumed that methane is the only hydrocarbon by-product released, but this is almost certainly an oversimplification; and if a substantial proportion of higher hydrocarbons is expelled, then estimated methane yields must be diminished proportionately. For example, in the artificial coalification experiments conducted by Saxby *et al.*, (1986), a large proportion of the volatile by-products of coalification were as hydrocarbon gases C₂ through C₅, plus a significant amount of solvent extractable oils and asphaltenes. (These products were produced at the end of year 4 of the experiment, when experiment temperatures had reached 300°C and vitrinite reflectance had reached around 1.0%). Concurrent production of methane was very small—around 9 cm³ (STP) per gram of solid product. Mass balance calculations indicate that if methane had been the only hydrocarbon product released, methane generation would have totaled close to 60 cm³/g.

Although the composition of the gases expelled under these experimental conditions (300°C) may not be representative of those that would have been formed and released during natural coalification (which proceeds at lower temperatures for longer periods of time) the observed differences in methane production figures may be of critical importance from the standpoint of reservoir behavior. The small amount of methane actually produced in the experiments would be barely sufficient to fully saturate a typical coal at this rank, whereas the larger amount would produce considerable excess gas which would

both saturate the reservoir and migrate into surrounding strata.

Most previously published estimates of methane generation during coalification have been made with respect to subhydrous, vitrinite-rich coals of Carboniferous age (e.g. Juntgen and Karweil, 1966; Mott, 1943). Coals of different geologic ages and differing petrographic compositions will behave considerably differently as hydrocarbon source rocks; and small differences in initial H/C ratio can give rise to significant differences in hydrocarbon generation potential (Saxby and Shibaoka, 1986; Higgs, 1986; Fig. 5). For example, inertinite-rich Gondwana coals, typical of some basins in Australia, are already depleted in oxygen and hydrogen even at very low rank. Accordingly, they cannot be expected to produce large quantities of additional hydrocarbons during maturation (est. 0-2%, by weight: Saxby and Shibaoka, 1986); and, as noted above, an oil-prone kerogen may liberate its hydrocarbons too quickly, in the form of oil. Hypothetically, certain varieties of vitrinite-rich coals may constitute the ideal source materials for methane generation. Such interrelationships need to be investigated more carefully.

The influence of grade on gas generation during organic metamorphism is apparently very simple, but perhaps deceptively so. Superficially, the less OM present, the less hydrocarbons that are formed. The true relationship may be more complicated, however, if mineral matter (particularly clay) influences the generation and/or expulsion of oils formed within the "oil window". A variety of research suggests that have indicated that clay minerals play an essential role in catalyzing the formation of oil during artificial pyrolysis experiments as well as influencing the process of expulsion (Saxby *et al.*, 1992). If oils formed during bituminization are able to escape from the coal structure prior to the onset of thermal gas generation, then there will be less hydrogen rich material available for conversion to methane at higher ranks. Any such relationships remain, however, well within the realm of speculation.

Hydrocarbon Retention

Coals at all ranks have the capacity to sorb methane within their structure, but this capacity changes substantially as a function of rank. Two rank-related compositional factors exert a fundamental control on methane sorption capacity: 1) the accessible "surface area", which is related both to the physical structure and chemical composition of the coal, and 2) competition with other species comprising the molecular fraction. It has been fairly routinely reported in the coalbed methane literature that methane sorption capacity progressively increases with rank (Kim, 1977; Eddy and Rightmire, 1982); but while this is partly true, the actual relationship is more complicated. Although there is quite a bit of scatter in the data, several studies have determined that methane sorption actually decreases with rank, passes through a minimum around 85% C (low volatile bituminous), then increases again at higher rank (Moffat and Weale, 1955; Jolly, 1968; Botham, cited in Patching, 1970; Schwarzer, 1983; Fig. 6).

In this respect, methane behaves identically to all other sorbates (discussed previously). Resolution of this apparent inconsistency is that the methane studies cited were all conducted on dry coals, whereas the coalbed methane data are generally based on measurements on moist coals. When water is allowed to compete for sorption sites within the coal structure, water usually "wins"—particularly for low rank coals. At low rank, water has the ability to displace methane from sorption sites within the coal structure, thus substantially diminishing the methane storage capacity of low rank (vitrinite-rich) coal. At high rank, however, coal retains its capacity for methane, while losing its affinity for water. Thus, the methane sorption capacity exhibits a net increase (Joubert *et al.*, 1974; Kim, 1977), but only for water-saturated vitrinite rich coals. The rank-related relationships for coals of other petrographic compositions may be quite different.

Interrelationships of sorbates other than methane and water are not yet well documented, but it is clear that to fully understand sorption behavior, the overall composition of the molecular fraction should be evaluated in its entirety. For example, stud-

ies of binary sorption behavior of San Juan basin coals have demonstrated a competition between nitrogen and methane at high pressure (Puri and Yee, 1990; Arri *et al.*, 1992). This characteristic was evaluated to test the feasibility of using nitrogen flooding as a commercial recovery method, but no rank related trends have yet been established. Binary sorption behavior of coals in CH₄-CO₂ systems have also been investigated (Olszewski *et al.*, 1992), but not yet in sufficient detail to draw any conclusions.

The influence of occluded oil within the coal structure on sorption capacity of two coals of similar rank is presented in Table 2 and Figure 4. Samples were collected of two Fruitland coals occurring within 100 ft (31m), stratigraphically, of one another in the same well drilled in the northwestern San Juan basin. Fixed carbon yield of the two was essentially identical, and vitrinite reflectance was similar. Nevertheless, the coals differed substantially in the composition of their molecular fraction, with the stratigraphically lower coal containing a much higher concentration of waxy oil as revealed by Rock-Eval pyrolysis and solvent (methylene chloride) extract studies. The waxy coal exhibited equilibrium moisture capacity and CO₂ surface area roughly half of that of the other coal and total gas yields measured by desorption testing showed roughly half as much gas desorbed from the waxy coal. Methane sorption capacity was also slightly lower in the waxy coal (Fig. 7). Apparently, the presence of waxy oil in the coal structure has diminished the coal's capacity to sorb methane, carbon dioxide and water.

It has been generally assumed that all underground coal seams are saturated with moisture, but this may not be the case. Some coal seams in the San Juan basin contain no free water in their fracture systems, and produce essentially no water during production (Decker and Secombe, 1986; Boyce and Levine, 1991). These coals may also be undersaturated with respect to their inherent moisture capacity, but this has not been proved. In any case, the composition of molecular constituents of coal cannot be assumed a priori and cannot be deduced from crude characterization methods such as proximate analysis. They should be directly

evaluated using modern organic geochemical methods.

The influence of different maceral "types" on sorption capacity is not well documented in the literature, particularly since there have been few studies where petrographic controls have even been considered. Most of the published data, both from North America and Europe, (e.g. Kim, 1977; Creedy, 1988) has been biased toward high vitrinite, Carboniferous age coals; and published results have failed to provide a clear picture of relationships in the reservoir.

Several studies in the Russian literature report influences of coal "type" on methane capacity of coals. Specifically, gas yields are reported to increase with fusain content, and decrease with increasing concentration of [some types of?] vitrain (Ettinger, I.L., 1966). Zabigaylo *et al.* (1972) indicate that, with rare exceptions, higher concentrations of structured gelified vitrain components (band-type vitrinite?) improve the sorptive capacities of coal, but they also report that fusain also has a positive influence. This same study showed that an increase in the proportion of fine-grained matrix [durain/desmocollinite?] decreases sorption capacity. Based upon multivariate statistical analysis of 57 desorption test cores from the Cahaba basin, Alabama, Levine (1987) discerned a weak positive correlation between inertinite content and gas yields, but inertinite concentrations varied very little in this data set. Faiz and Cook (1991) using similar methods reported a somewhat erratic but significant positive relationship between inertinite and gas yields for coals from the Southern coal field, N.S.W., Australia. After normalizing for other variables, an increase in inertinite content from 10 to 50% accounted for an increase in gas yield from around 6 to around 10 cm³/g. In a later paper, Faiz and Hutton (1992) suggested that lateral differences in gas composition were related to structural features; higher proportions of CO₂ were associated with anticlinal structures whereas CH₄ occurred in synclinal structures within the Permian Bulli seam. Creedy (1988) and Ulery (1988) found very little influence of petrographic composition on natural gas yields, but this may be reflective of the low variability in petrographic composition of their sample sets.

In contrast with these field-based studies, laboratory studies have shown vitrinite to have the highest sorption capacity of the three major maceral groups. For example, Thomas and Damberger (1976) showed that, over a range of ranks from high volatile C bituminous to high volatile A bituminous, vitrain bands had CO₂ surface areas that were on average 60 m²/g greater than "whole coal" samples of equivalent rank (Fig. 8). The petrographic compositions of the vitrain bands were not reported, but were presumably richer in "vitrinite A", or "band-type" vitrinite. A single sample of fusain (containing estimated 90% inertinite) had a CO₂ surface area of only 35 m²/g, as compared with 160 m²/g for whole coal samples and 220 m²/g for vitrain bands of the same rank. While it is not certain that this conclusion would also apply to methane, Jolly *et al.*, (1968) report significantly higher methane saturation capacities for bright bands (850 ft³/ton) as compared with dull bands from the same coal (570 ft³/ton), in spite of the two sub-samples having similar rank as measured by elemental carbon contents. This suggests that fundamental differences in the molecular structure of the two coal types may control sorption.

Lamberson and Bustin (1992) utilized a combination of high pressure methane sorption isotherms and low pressure CO₂ surface area studies to evaluate the influence of petrographic composition on coals from western Canada. Their results demonstrate that petrographic variability can exert at least as strong an influence on CO₂ and CH₄ sorption capacity as does rank. Total gas sorption varied directly with the proportion of vitrinite present (Fig. 9). There was also some evidence that band-type vitrinite (vitrinite A) had higher sorptive capacities than desmocollinite and vitrodetrinite (vitrinite B). These results need to be verified for other coals.

For the most part, mineral matter appears to serve merely as an inert diluent in sorption of methane in the reservoir. Correlation of desorbed gas yields vs. mineral matter content extrapolate to nearly zero gas content at 100% mineral matter (Levine *et al.*, 1990; Mavor *et al.*, 1991; Fig. 10). It should be noted, however, that the strong secondary fluorescence of clays in coals of the San Juan

basin suggest that they have adsorbed or otherwise retained significant amounts of oil generated in the oil window.

Migration

Economic production of natural gas from coal requires that methane and associated gases be able to migrate from the coal matrix where they are concentrated, to a production well from which they are produced. Transport through the reservoir is generally modeled as comprising two distinct steps: some sort of diffusion process by which the methane molecules "wriggle" through the tortuous macromolecular matrix of the coal into a relatively open fracture, followed by fluid flow through an interconnected network of fractures (Giron *et al.*, 1984). Gamson and Beamish (1991) suggested that the occurrence and orientation of coal phyteral porosity (stemming from remnant cellular structure) and cavities between grains in the attrital matrix, contribute to effective diffusion rates and pathways. Both steps must be considered in evaluating the influence of coal composition on reservoir production.

The diffusion step of gas transport is often modeled as Fickian diffusion (i.e. migration of molecules down a concentration gradient), but probably involves the combined influence of several distinct processes (Sevenster, 1959; King and Ertekin, 1989). Evidence from gas sorption studies suggests that the conduits through which gas molecules pass during diffusion are on the same order of size as the effective diameter of the molecules themselves (ca. 0.4-0.5 nm). Thus, coal behaves as a "molecular sieve", permitting some molecules to pass, while excluding others. Molecules migrate at different rates, depending on their size, but also depending on the molecular structure of the coal. Methane diffuses readily and higher hydrocarbons more slowly. Diffusion through coal appears to be an "activated" process, whereby a molecule must surpass a certain minimum activation energy in order to pass through the system (Nandi and Walker, 1966). For this reason, molecules migrate more readily at higher temperature, where a larger proportion of molecules possess the minimum energy required for transport.

The rate at which gas may migrate out of an unconfined coal sample (for example in a

core desorption test) is influenced both by diffusion rate and by fracture spacing (Hofer *et al.*, 1966; Kissell and Deul, 1974; Banerjee, 1988). Once a gas molecule has diffused into a fracture, it has effectively escaped from the coal, and its egress to the external boundary of the core is then very rapid. In the reservoir, where the fractures are more compressed owing to in situ stresses, the situation is likely to be more complex, but actual production behavior of wells demonstrates that fracture (i.e. cleat) development is still an important factor influencing production (e.g. Mavor *et al.*, 1991).

By whatever combination of mechanisms, however, the concentration of occluded gas responds much more rapidly for some coals than for others to changes in ambient conditions. To a large degree these differences are related to coal composition. It can be difficult to distinguish, however, if diffusion rate or path length (i.e. fracture spacing) is the important rate-limiting factor unless these characteristics have been directly measured.

Coal rank appears to have a direct relationship both to diffusion rates and to fracture spacing. Coals near the peak of bitumization require higher activation energies for gas migration than coals of lower and higher rank (Fig. 11). Based upon the results of core desorption tests, the same relationship appears to hold for methane diffusion as well. The percentage of so-called "residual", or "remaining" gas measured in core desorption tests (Diamond and Levine, 1981) passes through a maximum at high volatile A bituminous rank (Fig. 12). The term "residual gas" pertains to volatilizable constituents that remain entrapped within the coal structure even after extended periods (several weeks or months) of desorption at room temperature and pressure. The rate of release of this residual component can be increased substantially by comminuting the coal to a fine micron-sized particles in a hermetically sealed ball mill (Diamond and Levine, 1981), suggesting that diffusion is the rate-limiting process. This slow release rate is apparently a consequence of high volatile A coals having lower diffusivities than coals of higher and lower rank, possibly owing to obstruction of flow paths by occluded hydrocarbons. In one case, a sample of high volatile A coal (Pittsburgh coal bed) has continued to release

small amounts of desorbed gases even after several years of continuous desorption (J. Ulery, 1991, personal comm.).

Cleat spacing is known to be related to coal rank, but stress regime during maturation and lithotype composition exert an influence as well. The spacing of fractures can vary from millimeter scale up to 10s of centimeters, and is apparently related to coal rank, bed thickness, and composition. Cleat is typically not well developed in most low rank coals, but becomes progressively better developed through the bituminous rank series. The closest cleat spacings are typically observed in coals around low volatile bituminous rank. Beyond low volatile bituminous rank cleat fractures appear to be rehealed, although shadowy remnants of former fracture surfaces are still discernible. Mechanisms for rehealing are not known, but might include either annealing or repolymerization of coal structure across fracture planes.

At any given coal rank, cleat spacing is primarily a function of composition. Macrae and Lawson (1954) report that in the Yorkshire coal field durain bands thicker than 6 inches (15 cm) had fewer than 5 fractures per foot (0.2 per cm), while bright coal (vitrain & clarain) had as many as 70 fractures per foot (2 per cm). Likewise, dirty (clay rich) layers in coal seams are obviously less cleated than clean layers. Decker *et al.* (1989) report that cleat spacing is inversely proportional to ash yield (although the correlation is very poor) in coals from the Cedar Hill field, San Juan Basin. In Australian coals, Gamson and Beamish (1991) and other unpublished work have found similar correlations of increased cleat and microfracture frequency and density in the bright vitrain bands than in the finer grained attrital matrix, or dull bands.

Qualitatively, denser, harder coals seem to desorb gas more slowly than more friable coals (McCulloch *et al.*, 1974, 1975), but this may be more a function of fracture spacing than of diffusion rates. Studies of re-equilibration rates of powdered samples show (somewhat unexpectedly) that high ash coals and carbonaceous shales actually equilibrate more rapidly to changes in ambient gas pressure than do clean (low mineral matter) coals. Similarly, analysis of a suite of core specimens from the Cahaba basin, Alabama

revealed that higher ash samples, although they contained far less gas than low ash samples, actually released entrapped gases more rapidly (Levine, unpubl. data). Far more work is required to fully elucidate these relationships.

CONCLUSIONS

In comparison with the advanced state of knowledge pertaining to conventional hydrocarbon reservoirs, comparatively little is known about coal as a reservoir rock. On the basis of available data, variation in coal composition accounts for a high degree of variability in the behavior of coal bed reservoirs. It is further apparent that coal beds are much more variable and complex than has been generally acknowledged in some of the early literature on this topic (Levine, in press). In this regard there is a very large "information gap" between the comparatively sophisticated state of knowledge of coal composition and molecular structure gained by coal chemists over the past two decades and the comparatively crude models of coal composition and structure utilized in coalbed methane exploration programs and reservoir modeling. Much will be learned by melding information from different disciplines. These problems demand an interdisciplinary approach, involving (at least) the combined expertise of sedimentologists, coal petrologists, organic geochemists, and reservoir engineers, among others.

Unfortunately much of the previous coal work is inconclusive owing to inadequate sampling procedures, erroneous measurement procedures, and/or data sets that are too small to yield statistically significant relationships. Comprehensive, broad-based studies are almost absent from the published literature, due in part to the additional cost and, therefore, low priority placed on detailed reservoir characterization.

Of particular significance in reservoir analysis is the recognition that coal is not accurately described as a solid reservoir rock with very tiny "pores", but rather as a complex mixture of different organic substances, including many that would be free liquids or gases were it not for their intimate association within the coal structure. Coal appears to

have a three-dimensional macromolecular matrix structure, but also includes a large proportion of the materials that are free molecules, held in place either by weak hydrogen bonding, van der Waals bonding, or physical entanglements. Owing to inefficiencies in the packing of this complex mixture, there is plenty of additional room for small molecules such as water and methane. When viewed as just one component in this multi-component mixture, "coalbed methane" seems to have a significantly more complex relationship in the reservoir than has been envisioned in the past.

Recent studies of coal as a source rock for oil have demonstrated that methane is but one of a diverse suite of hydrocarbon substances generated by coal during coalification. Different coals appear to behave quite differently in this regard, with some coals generating much more (or less) methane than others. Specifically, coals which expel significant amounts of oil must produce proportionately less methane; and hydrogen-poor inertinite-rich coals have very low generative potential.

Debates regarding the comparative methane sorptive capacities of different coal or maceral types are clouded by inconsistent results. Several laboratory studies appear to show vitrinite as having considerably higher sorbate accessibilities than inertinites, yet field data appear to show the opposite relationship. It is possible that the difference may be due to the measurement conditions, with the presence (or absence) of water playing a role. Sorbed oil also appears to suppress methane sorptive capacity, but available data are very scanty. Such speculative interrelationships can be resolved only with additional study.

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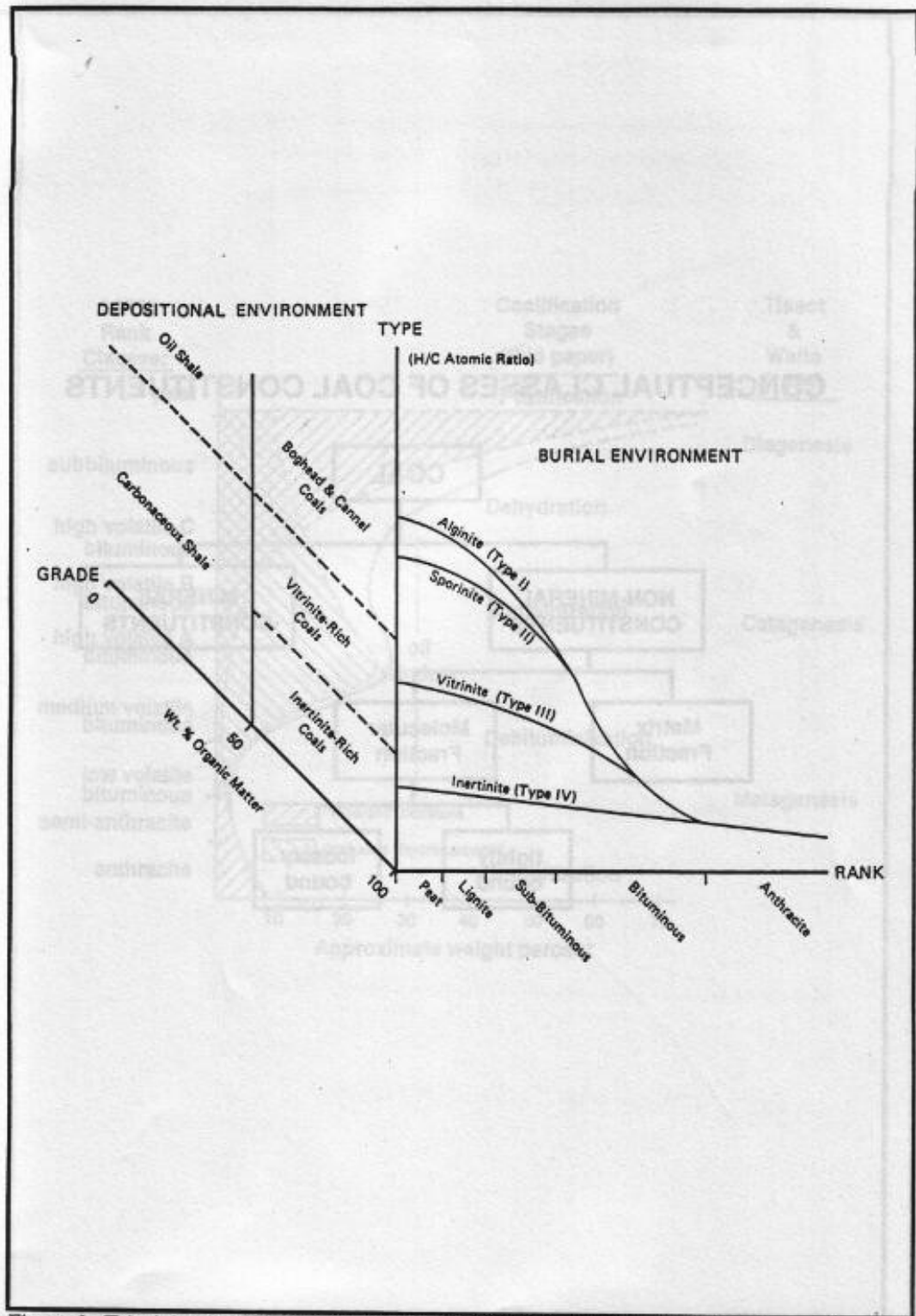


Figure 1. Triaxial diagram representing all varieties of organic matter-bearing sedimentary rocks.

CONCEPTUAL CLASSES OF COAL CONSTITUENTS

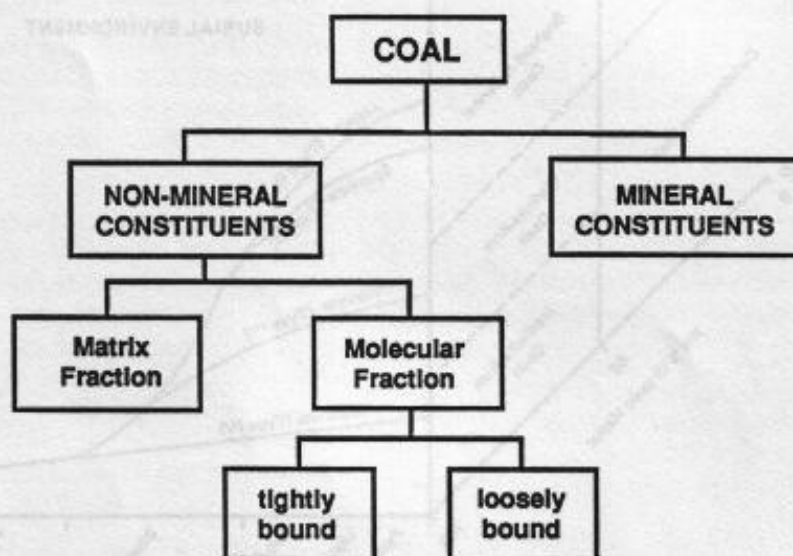


Figure 2. Conceptual classes of coal constituents, including matrix and molecular fractions

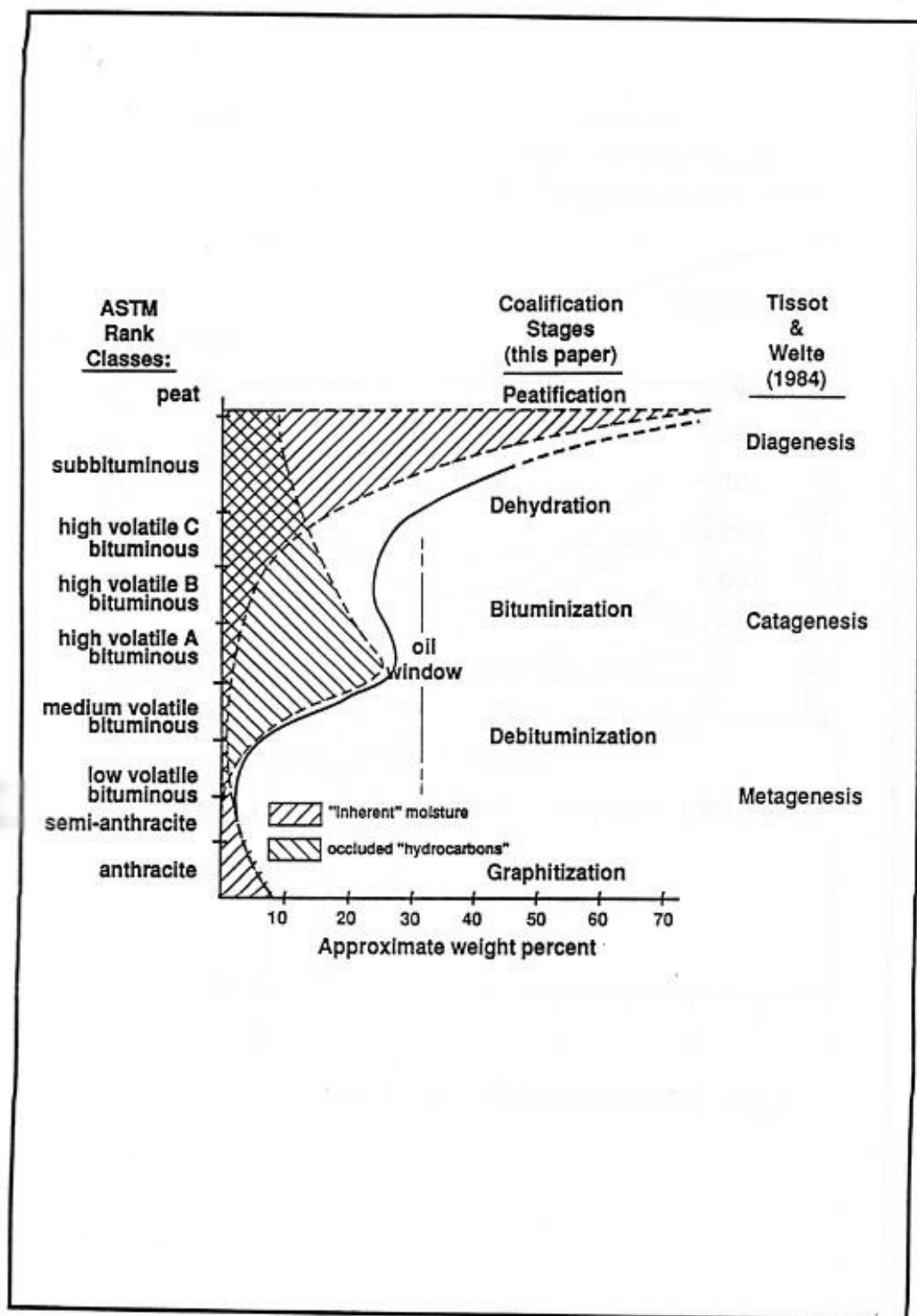


Figure 3. Evolution of the molecular fraction of coal during coalification (after Levine, in preparation).

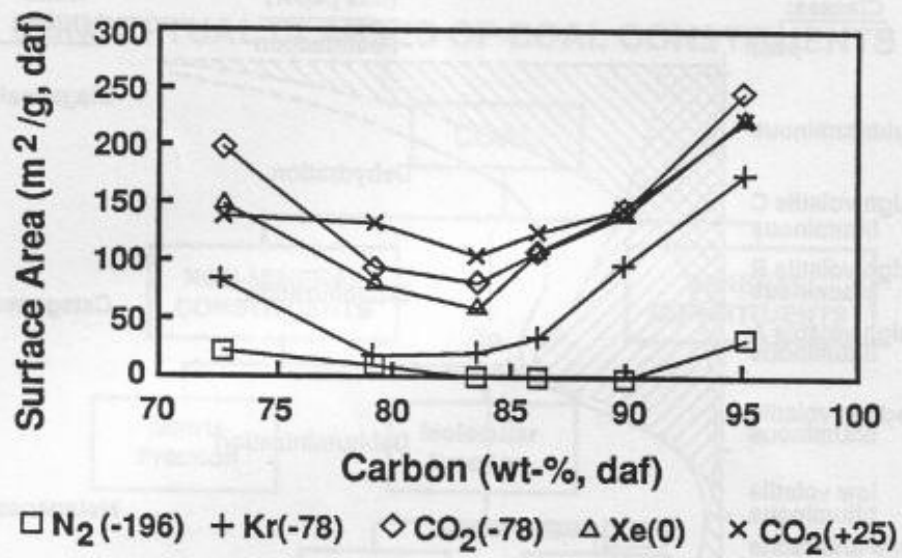


Figure 4. Change in surface area with rank for different sorbates (from Walker and Kini, 1965).

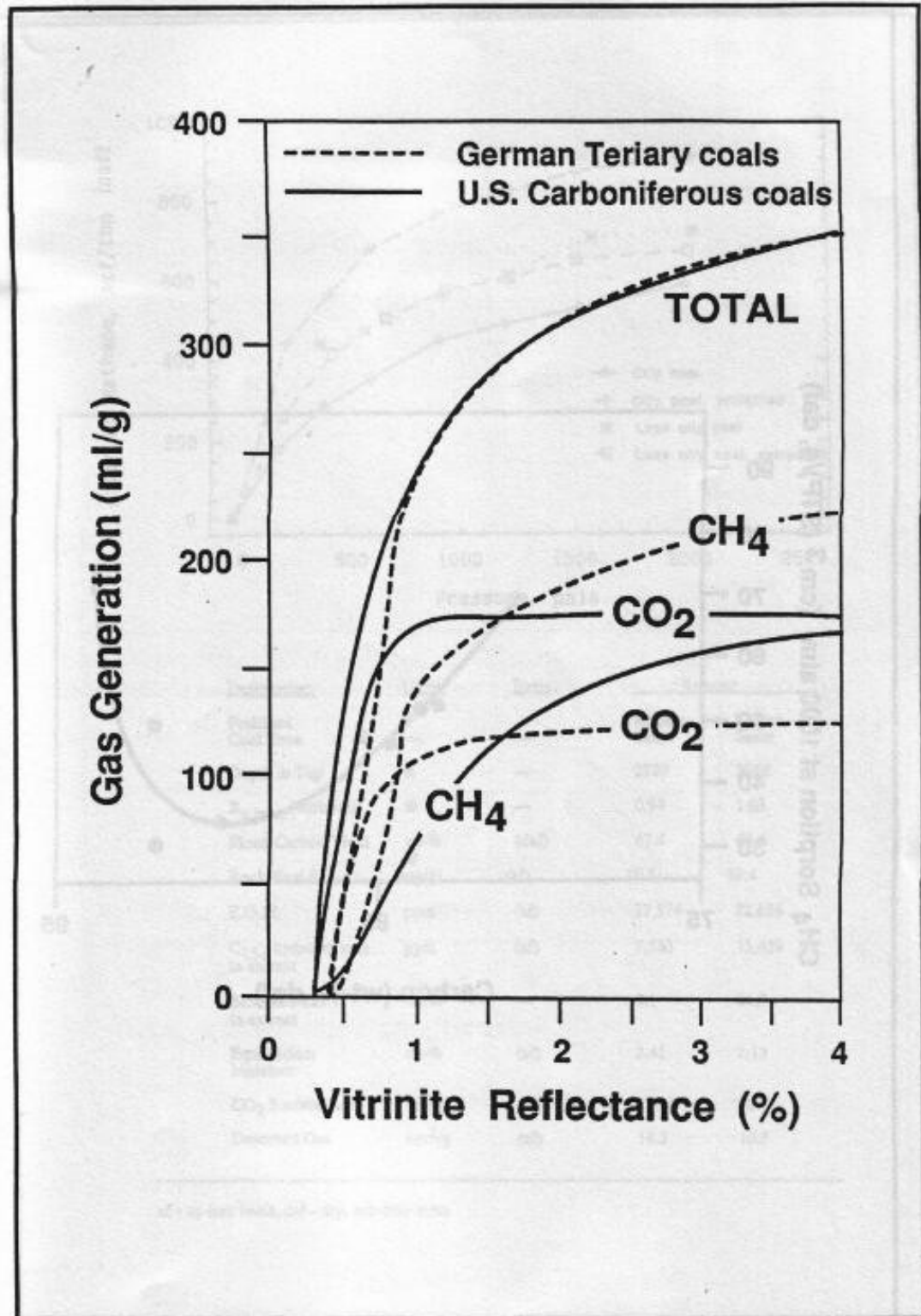


Figure 5. Cumulative methane generation curves for two broadly differing coal types: a perhydrous German Tertiary coal and a subhydrous U.S. Carboniferous coal (after Higgs, 1986).

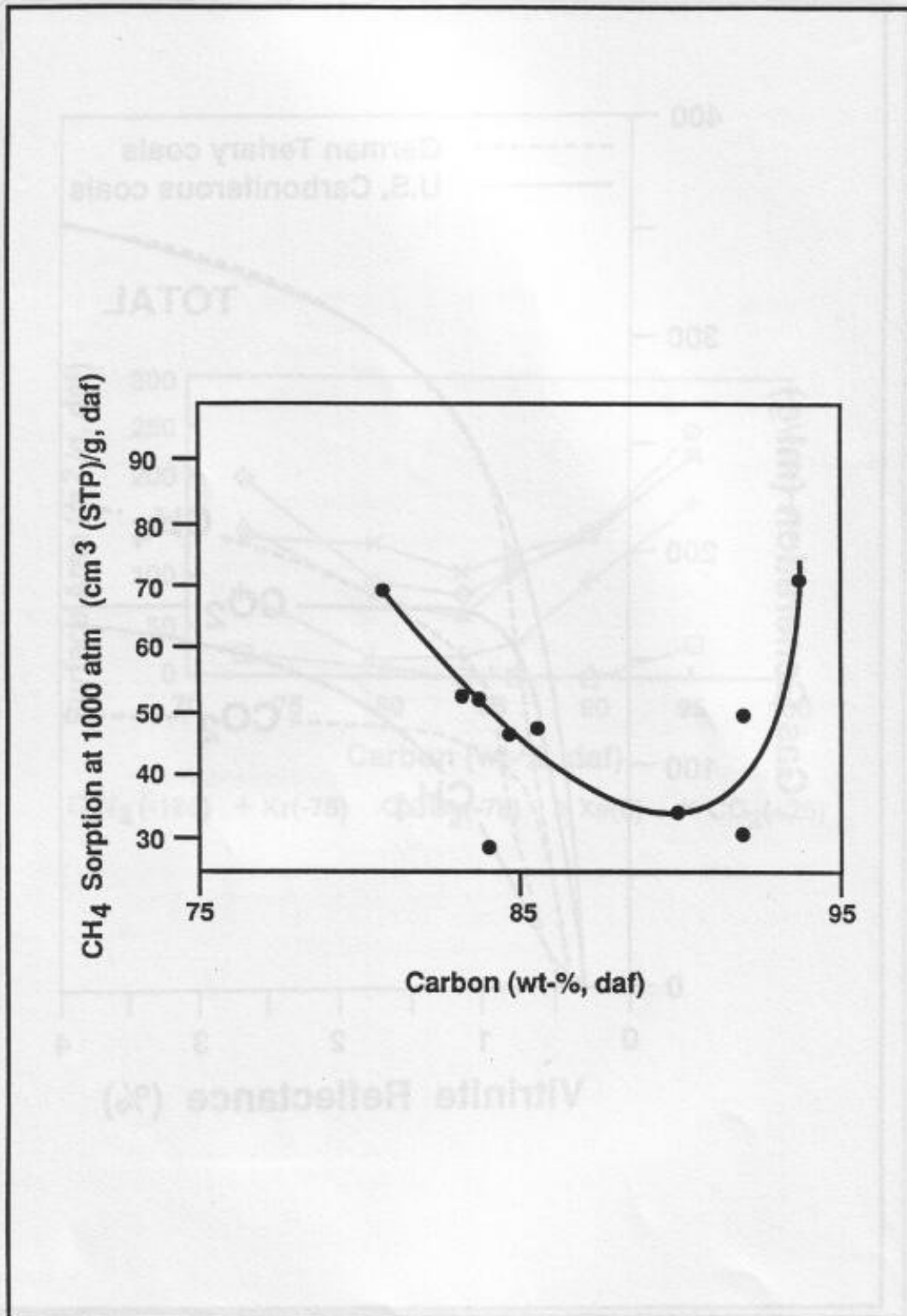


Figure 6. Methane sorption capacity as a function of coal rank (after Moffat and Weale, 1955).

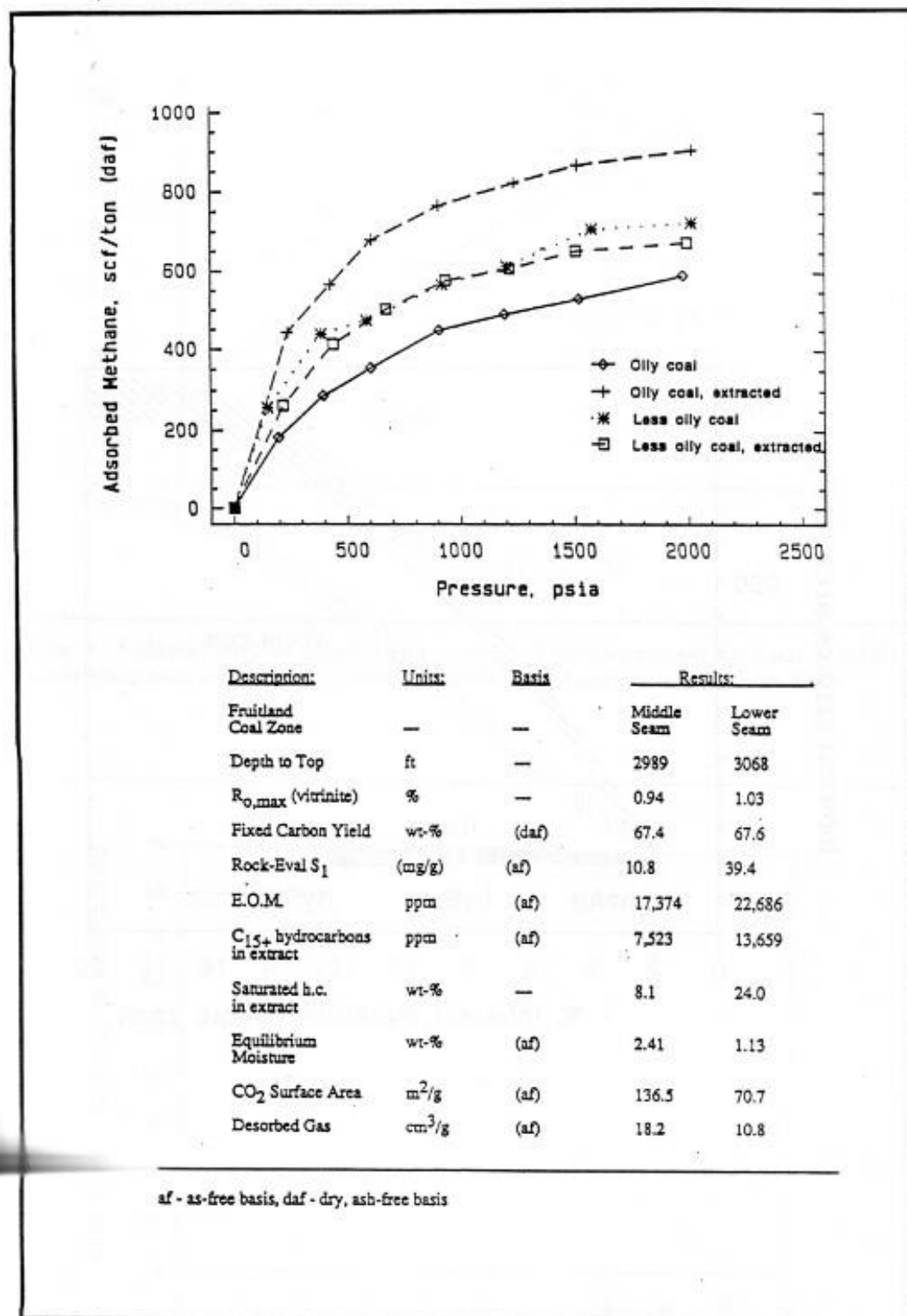


Figure 7. Methane sorption isotherms at 30°C for two stratigraphically nearby coal seams in the Fruitland coal group, San Juan basin, Colorado. The coal higher in occluded waxy hydrocarbons (lower seam) sorbs smaller amounts of methane in its natural state than the middle coal, but after extraction with methylene chloride, sorbs significantly more. The accompanying analytical data for the two samples shows the differences in composition and sorbate accessibilities (from Levine, in press).

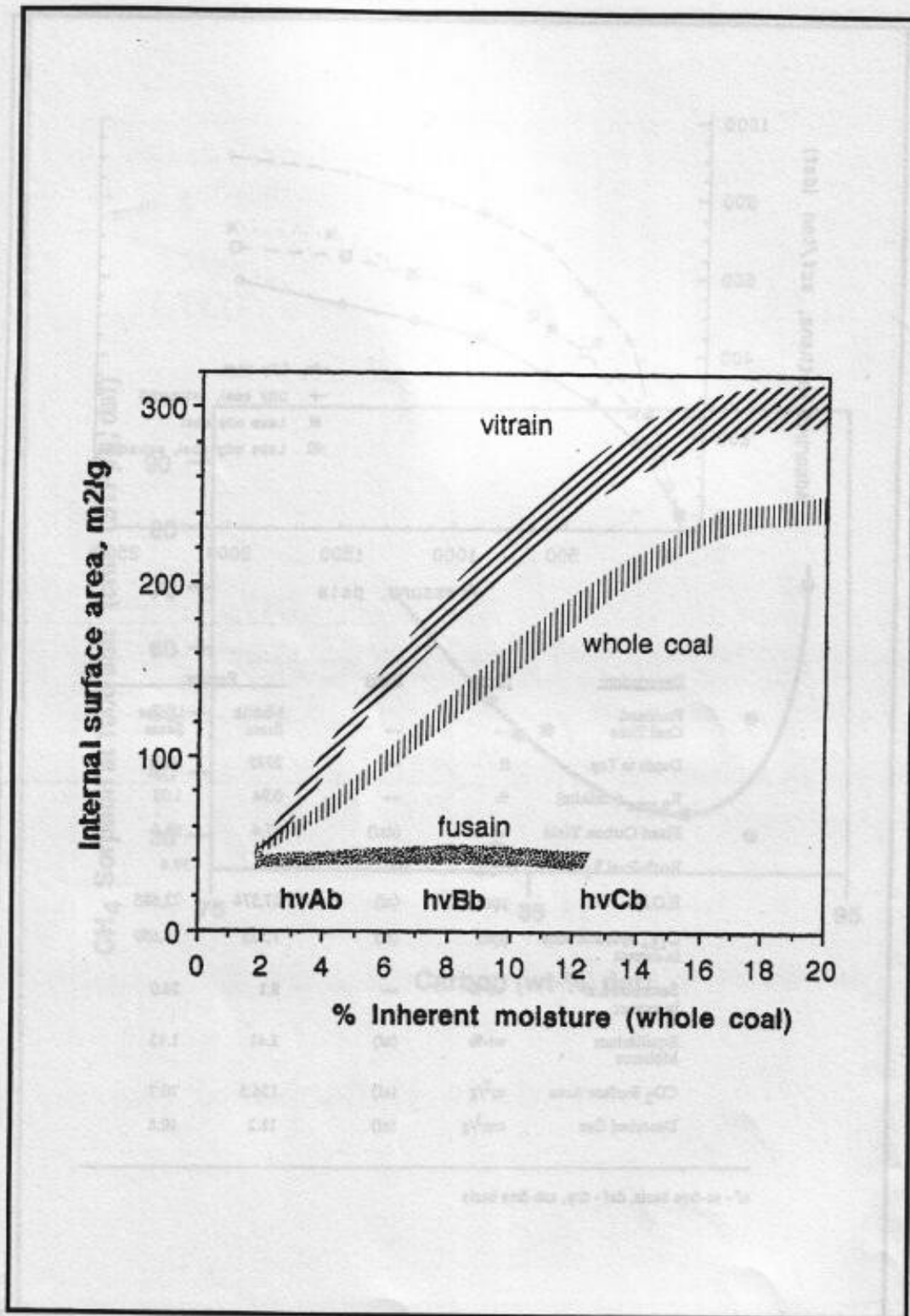


Figure 8. Variation in CO₂ surface area as a function of coal rank for whole coal samples and vitrain concentrates from the Illinois basin. Coals range in rank from high volatile C bituminous to high volatile A bituminous (after Thomas and Damberger, 1976).

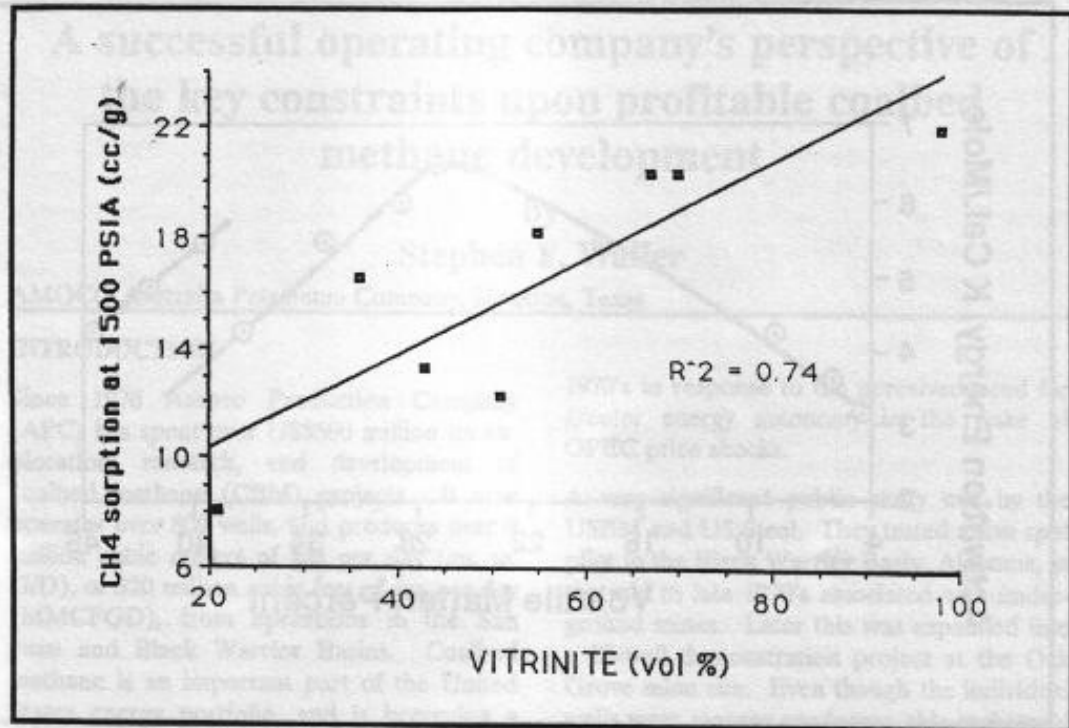


Figure 9. Methane sorption capacity as a function of vitrinite content for coals of medium volatile bituminous rank, British Columbia, Canada (from Lamberson and Bustin, 1992).

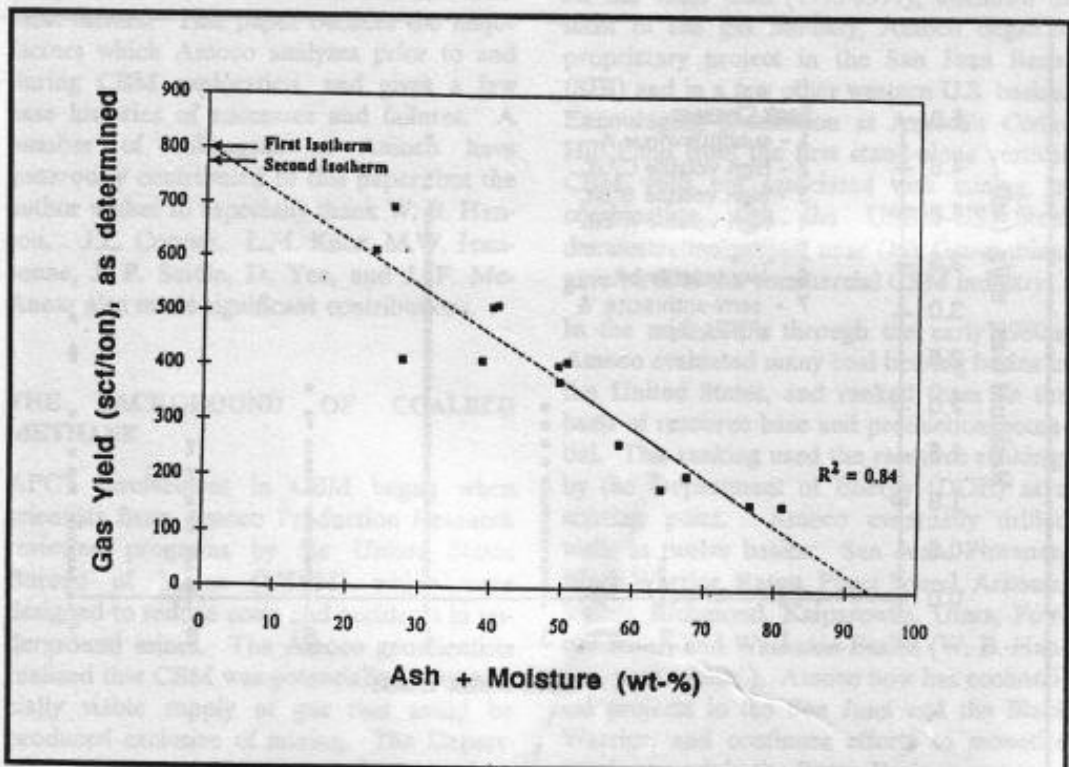


Figure 10. Natural gas yields of test cores from the San Juan basin, Colorado, showing the negligible contribution of ash-forming minerals and water to total sorption (Mavor *et al.*, 1991).

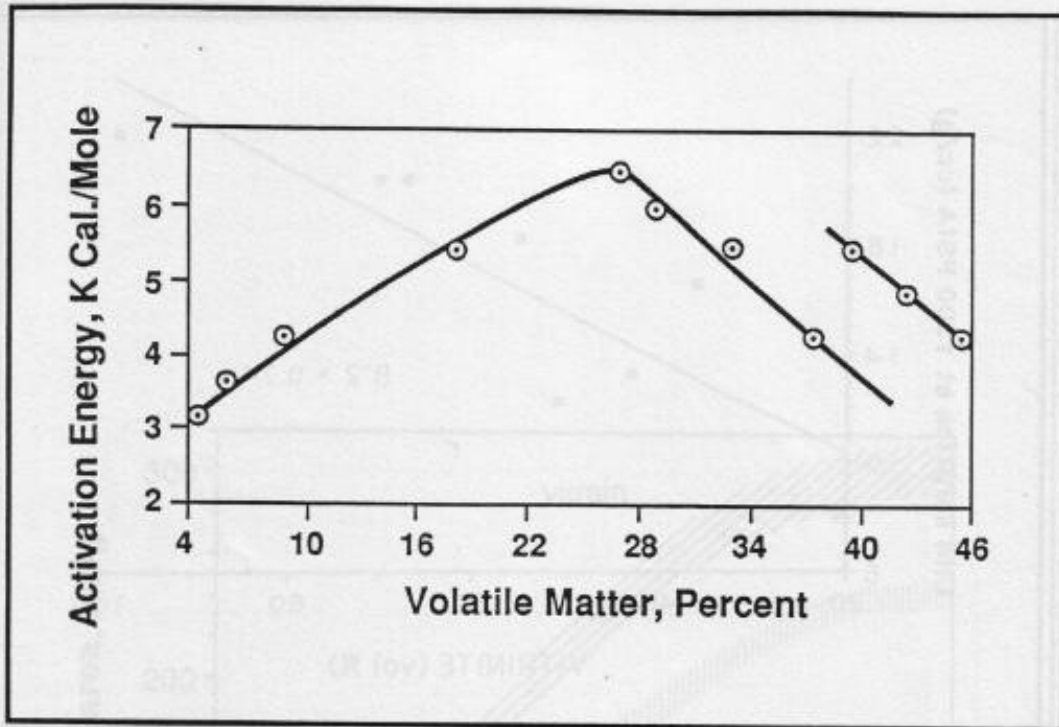


Figure 11. Activation energies for Argon diffusion as a function of coal rank (after Nandi and Walker, 1966) >

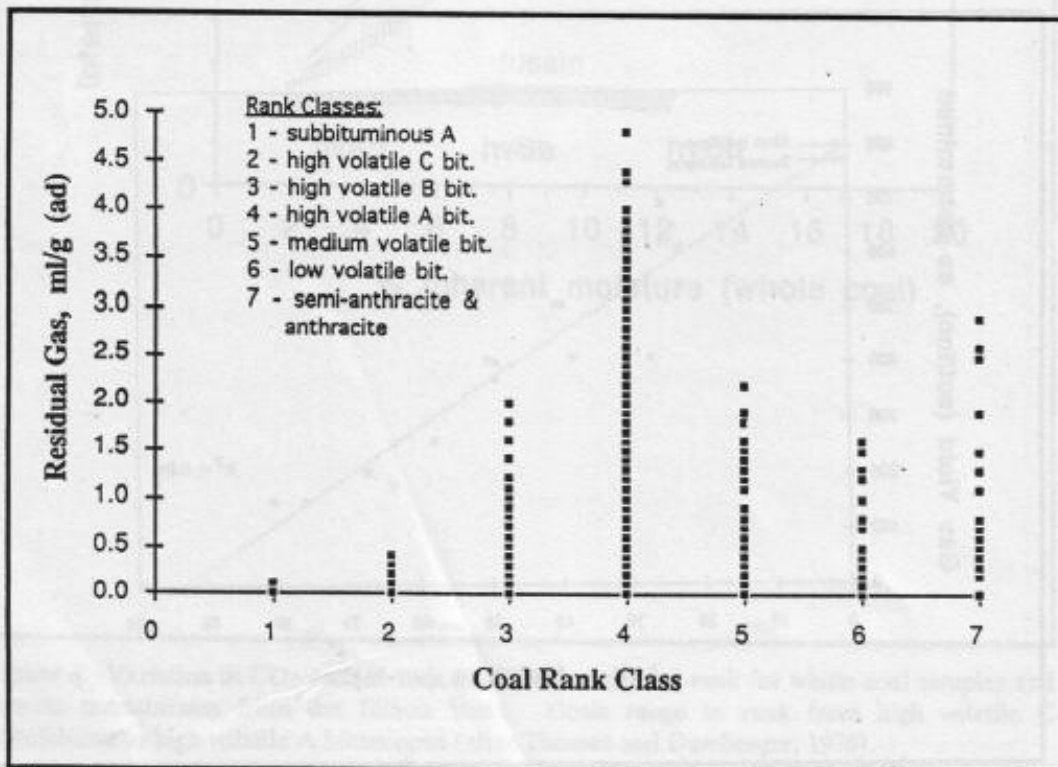


Figure 12. Residual gas as a function of coal rank (based upon data reported in Diamond and Levine, 1981).