# Dissolved Organic Constituents in Coal-Associated Waters and Implications for Human and Ecosystem Health

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## ABSTRACT

Decisions concerning the disposal or use of waters produced during the production of coal bed methane (CBM) are typically made based on water quality as expressed by the water's salinity and/or inorganic ionic constituency. The quality of CBM water is frequently evaluated in terms of its suitability for irrigation. Water that has an acceptable salinity and sodium adsorption ratio (SAR) is considered safe for surface discharge and for potential injection into a drinking-water aquifer. It is important to remember, however, that water associated with coal seams, independent of its inorganic constituents, may contain dissolved organics and other constituents at levels that may adversely affect human health and the environment. It is well known that coal, lignite, or coaly materials present in aquifers used as drinking-water supplies are associated with adverse or potentially adverse human health effects. Water produced from coal-associated aquifers has been linked, or is suspected to be linked, to goiter, Balkan Endemic Nephropathy (BEN), multiple sclerosis, and increased rates of cancer morbidity and mortality. Water-soluble organic compounds found in coals include goitrogens such as the hydroxyphenols resorcinol, 2-methyl resorcinol, and 5-methylresorcinol (orcinol), as well as hydroxypyridines. Well waters containing polycyclic aromatic hydrocarbons (PAHs), aniline, aminophenols, and aromatic amines leached from low-rank Pliocene coals may be the cause of, or a contributing factor to, BEN, an incurable interstitial nephropathy that is believed to have killed more than 100,000 people in the former Yugoslavia alone. There are few, if any, systematic and comprehensive analyses of dissolved organic compounds in CBM-produced water. Prudence suggests that the dissolved organic constituents in CBM-produced water should be systematically characterized, and their potential for harm to human health and the environment be evaluated before potentially harmful chemicals are discharged to the environment or released to drinking-water aquifers.

# INTRODUCTION

## Coal Bed Methane

The term "coal bed methane" (CBM) refers to gas that is generated during coalification and stored within coal on internal surfaces. This gas can be generated by microbial processes but is principally generated through thermal decomposition of the coal. CBM is generally referred to as an unconventional source of fossil fuel; however, it is a significant source of methane in the U.S. and worldwide. In 1999, proven reserves of CMB in the conterminous U.S. was about 13.2 trillion cubic feet (Tcf) (~7.5% of total U.S. proven reserves), and the annual U.S. production of CBM was about 1.25 Tcf (~6.3% of U.S.-associated and non-U.S.-associated gas production) (Energy Information Administration 2000). Worldwide estimates of in-place resources are as much as 7,500 Tcf, but the scarcity of basic data on coal resources and gas content makes this number uncertain (Rice 1997).

CBM is a more environmentally acceptable energy source than that derived from the burning of coal because the production of CBM does not produce the environmental damage of coal mining or waste ash and sulfur gases that result from coal combustion. Further, CBM resources are widespread, and, because coal can store 6 to 7 times more gas than the equivalent rock volume of a conventional gas reservoir, are characterized by large in-place resources (Rice 1997). An additional attraction of CBM development is the low cost of drilling and completing CBM wells. Often, substantial CBM resources can be reached at very shallow depths, and because of the often spatially broad occurrence of suitable coals, exploration costs for CBM development are minimal.

The initial focus of CBM development in the U.S. was the Black Warrior basin in Alabama. This focus shifted in the late 80s and early 90s to the San Juan Basin in New Mexico and Colorado. In 1995, approximately 94% of CBM production in the United States was from the San Juan Basin in New Mexico and Colorado and the Black Warrior Basin in Alabama (Stevens et al. 1996). Today, although New Mexico, Colorado, and Alabama still account for 90% of CBM production, the focus of CBM development is the Powder River Basin in Wyoming and Montana. At present, there are approximately 3,000 CBM wells in the Wyoming portion of the Powder River Basin; however, planners forecast a staggering number of new wells: under what is described as a "moderate" scenario, a total of 81,000 CBM wells are projected for Wyoming, with 50,000 of these drilled by 2010 (Bureau of Land Management 2001). Based on recent proposals by operators, about 9,500 CBM wells will be drilled in the Montana portion of the Powder River Basin by 2010 (Energy Information Administration 2000). The projected 90,500 new CBM wells in the Powder River Basin would constitute a 29% increase from the 307,449 wells producing gas and gas condensate operated in the U.S. in 1999 (Energy Information Administration 2000). CBM is also produced in Oklahoma, Virginia, West Virginia, and Pennsylvania.

The chief environmental concerns from CBM production arise from: 1) the requirement to dispose of large volumes of produced water, 2) the potential for the uncontrolled release of gas from the coal reservoir to shallow groundwater, 3) the

potential for drawdown of shallow groundwater and, 4) the potential for certain well completion technologies to affect shallow groundwater.

The rapid development of CBM in the U.S., the short experience with this type of gas production, and the potential or actual environmental impacts have produced an adversarial climate between CBM developers, surface owners, surface resource users, and environmental groups.

## Disposal and Use of CBM-Produced Water

Compared to conventional natural gas production, CBM production is accompanied by an enormous amount of water. For example, in 1990, CBM water production in the U.S. was 61 million barrels (MMbbl). This volume constituted 15% of the conterminous onshore water production associated with gas in 1990 (Gas Research Institute 1995). Because water production in CBM development is higher early in the life of a well, the greatest water production in CBM exploitation comes early in the life of that development. The amount of water produced by a CBM well and the ratio of gas to water in the produced fluid depends on many factors, including the duration of CBM production, original depositional environment, depth of burial, and type of coal. Water production statistics for some major CBM-producing basins in the U.S. are presented in Table 1. Note that in all instances the water/gas ratio for CBM is high (bbl/Mcf 0.03–0.1) to very high (bbl/Mcf >0.1) according to a categorization of ratios of produced water to produced gas published by the Gas Research Institute (1995).

The volume of water co-produced with CBM creates an enormous disposal problem. Depending upon local regulations and the salinity of the water, CBM waters are disposed of by injection into geologic formations, through evaporation and/or percolation in disposal pits, road spreading, and surface discharge. Except for injection into deep geologic formations, all other disposal options carry a risk of contamination of shallow groundwaters, and obviously, surface discharge of CBM water may result in the contamination of surface waters. Evaluations of the quality of CBM water have focused on its salinity and its balance of cations while the organic constituency of CBM water has been largely ignored. Because water associated with coal, especially water associated with lignites and subbituminous coal, is known to contain a wide variety of toxic or potentially toxic organics, the organic constituency of CBM waters should be thoroughly understood and evaluated during the process of selecting disposal options.

# COAL CHEMISTRY AND GROUNDWATER

## Coalification

The process of converting living biomass that has accumulated *in situ* into coal is termed coalification. In the diagenetic phase of this process, dead plant material (peat) undergoes biochemical transformation by microorganisms, chemical reaction, and physical compaction. The volume of the peat is reduced (i.e., its density increases), its water content is decreased, and its chemistry is modified. The primary plant structural biopolymers cellulose and lignin are both affected in that cellulose is largely degraded and lignin is chemically modified. The most noticeable gross chemical change during diagenesis is the differential loss of oxygen (as well as hydrogen and nitrogen) from the

peat with respect to carbon. In addition, the ratio of aromatic carbons (carbon atoms associated with graphite-type structures) to aliphatic carbons (carbon atoms associated with saturated chain and ring structures) increases. As burial depth increases, temperature increases, and at some point, biochemical activity ceases, and there is no further oxygen input. This phase of coalification is termed catagenesis, and is purely a thermochemical process. During the catagenic phase, oxygen and nitrogen continue to be lost and the aromatic character of the residual organic matter increases. In addition, the residual organic matter continues to be reorganized through both polymerization and depolymerization reactions. In summary, as coalification proceeds, the chemical structure of the coal becomes increasingly aromatic, richer in carbon and depleted in oxygen, nitrogen, and hydrogen.

For purposes of commercial (but not chemical) classification, coaly materials are assigned a "rank" based in part on the degree to which the coalification process has proceeded (ASTM 1995). The least mature coals are termed lignites and the most mature coals are termed anthracites. Table 2 provides summary information concerning the carbon, oxygen, hydrogen, and nitrogen content and the gross visual appearance of coals of various ranks. Although a gross correlation exists between coal rank, carbon content, and oxygen, the relationship between coal rank and hydrogen and nitrogen content is less apparent. Further, the ranges of carbon, oxygen, hydrogen, and nitrogen content reported overlap between successive ranks. Even though coal rank cannot be viewed as connoting a clear vision of coal chemistry, the term and concept are widely used, and, therefore, offer some utility when attempting to estimate the chemistry of coals from field or commercial descriptions.

## Chemical Structure

The chemical structural outcome of coalification is the conversion of cellulose (Figure 1), lignin (Figure 2), and other plant biochemical components to a large matrix of aromatic clusters connected by aliphatic bridges that possess aliphatic and carbonyl side chain attachments and some weakly bonded small molecular components (sometimes referred to as the mobile phase) (Smith et al. 1994; Given 1986; Krevelen 1981). A schematic view of coal structure is provided in Figure 3. The aromatic clusters consist largely of carbon, but also contain heteroatoms such as oxygen, sulfur, and nitrogen. The bridges connecting the aromatic clusters are generally believed to be almost exclusively composed of aliphatic functional groups, but may also contain atoms such as oxygen and sulfur (Solomon 1981). Bridges containing oxygen as ethers have relatively weak bond strengths. Some bridges, known as char links, consist of a single bond between aromatic clusters that are thought to be relatively stable.

## Organic Geochemistry of Groundwater in Coaly Aquifers

The presence of coaly material within an aquifer will modify the organic geochemistry of groundwater within that aquifer. Thermal stress (accompanying burial) and chemical oxidation (during burial or as the consequence of the introduction of oxygenated water or air) will cleave some of the bridge structures binding together the aromatic structures within the coal matrix. The cleavage of these bridge structures in the coal matrix can produce water-soluble organics, and it is well known that microorganisms can solubilize coals to a limited extent (Klein 2001). Because coals of lower rank contain a greater abundance of more easily cleaved bridge structures than

coals of higher rank, solubilization of lignite by microorganisms is more extensive than solubilization of bituminous coal, and the volatile matter and oxygen content of the coal are the principal factors influencing the extent of bioconversion (Reiss 1992). Consequently, it is reasonable to conclude generally that groundwater associated with coals of lower rank should contain higher levels of dissolved organics derived from the coal than groundwater associated with coals of higher rank, and that coals containing more volatiles and more oxygen should yield more dissolved organics. By inspection of the chemical structure of coal, the anticipated water-soluble organics should include a wide variety of oxygen-bearing aromatic compounds (e.g., phenols, aldehydes, ketones, and various carboxy-, hydroxy- and methoxy- bearing compounds), nitrogen-bearing compounds (pyridines and amines), mono- and polycyclic aromatic hydrocarbons (PAHs), and to a small degree, aliphatic compounds.

## Wyodak-Anderson Coal

The Wyodak-Anderson coal zone in the Powder River Basin in Wyoming and Montana is a low-sulfur coal of subbituminous rank. Based on the Penn State Coal Database[1], the carbon, oxygen, hydrogen nitrogen and sulfur composition of the Wyodak-Anderson coal is given in Table 3. These data demonstrate that the Wyodak-Anderson coal in the Powder River Basin in Wyoming and Montana is, compositionally, a typical subbituminous coal. That is to say, it possesses a substantial content of both oxygen and volatile matter. From the foregoing discussion, the Wyodak-Anderson coal would be expected to yield a substantial concentration of water-soluble organics in associated groundwater if it is exposed to thermal stress or oxidation reactions.

# HEALTH ISSUES AND COAL-ASSOCIATED GROUNDWATER

The suite of water-soluble organics anticipated to be present in groundwater associated with coaly material, especially lignite or low-rank coal, is expected to be largely aromatic in character and to contain oxygen- and nitrogen-bearing functional groups. This suite of materials includes numerous compounds that are toxic and/or carcinogenic. The limited number of studies concerning dissolved organics in groundwater associated with coal indicates that there may be an association between coal-associated groundwater and two diseases: endemic goiter and Balkan Endemic Nephropathy (BEN). Some investigators have also reported the occurrence of multiple sclerosis clusters in geographic areas that have drinking-water-associated peat and organic-rich rocks. There is also limited epidemiological evidence (spatial correlation) for an association between digestive cancers and the probable presence of coal in aquifers in Missouri (Kakey et al. 1980).

## Goiter

[1] Penn State Coal Sample Bank and Database available at http://www.ems.psu.edu/COPL/coal.html, downloaded August 22, 2002; values for carbon, oxygen, hydrogen and nitrogen are dry, mineral matter free weight %.

Goiter, which is also known as Grave's Disease, is a condition involving the enlargement of the thyroid gland and is generally not associated with inflammation or cancer. The thyroid gland may be two to three times the normal size as a result of antibodies called thyroid-stimulating autoantibodies that stimulate increased cellular proliferation. Individuals with goiter have increased basal metabolic rates, weight loss, muscle weakness and extreme fatigue, paradoxically with greatly reduced levels of thyroid stimulating hormone (TSH). Severe cases of goiter are manifested by mental retardation, decreased fertility, and infant mortality. Treatment of goiter typically involves the removal of the thyroid gland or administration of radioactive iodine to reduce thyroid activity and cellular proliferation. About 400 million people worldwide are affected by endemic goiter, which occurs in groups of people living within specific geographic areas.

The main cause of endemic goiter is an insufficient dietary supply of iodine. Typically, endemic goiter is related to iodine-depleted soil, and the use of iodized table salt often prevents the occurrence of these goiters. However, naturally occurring goitrogens have been documented in the case of certain foods (milk, cassava, millet, nuts, cabbage) and bacterial and chemical water pollutants (Gaitan 1983; Delange 1994). The mechanism by which the thyroid gland adapts to an insufficient iodine supply is to increase the trapping of iodide, as well as the subsequent steps of the intrathyroidal metabolism of iodine leading to preferential synthesis and secretion of triiodothyronine. This metabolic process is maintained by increased secretion of TSH, which is ultimately responsible for the development of goiter.

Endemic goiter in iodine-sufficient areas of the U.S. (Owsley and Breathitt Counties, Kentucky) and western Columbia (Sibundoy Valley) has been reported to be associated with the consumption of groundwater containing goitrogens derived from shales and coals (Gaitan et al. 1989). Further, experimental studies have shown that aqueous extracts from coal and black shale significantly inhibit thyroid peroxidase (TPO)[2] in porcine thyroid slices (Lindsay et al. 1992). Chemical characterization of coal-derived organic pollutants in groundwater used for drinking from iodine-sufficient goiter areas identified the presence of hydroxybenzenes (phenol, resorcinol), phthalate esters, sulfurated organics (thiophenes, disulfides), PAHs (naphthalenes, methoxyanthracene), bromoform, and aliphatic hydrocarbons (Gaitan et al. 1989). Inhibition of TPO by the most abundant organic compounds found in aqueous effluents from a coal conversion process (the dihidroxyphenols resorcinol, 2-methylresorcinol, and 5-methylresorcinol) and hydroxypyridines (hydroxypyridines and dimethyl- and ethyl- substituted hydroxypyridines) is comparable to or greater than the antithyroid drug 6-propyluracil (PTU) (Lindsay et al. 1992).

## Balkan Endemic Nephropathy

BEN is a fatal kidney disease that occurs in geographically discrete areas of the Balkan Peninsula (Serbia, Bosnia, Croatia, Bulgaria, and Romania) (Ceovic 1992; Plestina 1992). The disease was first described in 1956, but may have existed for many centuries (Tanchev 1991). Individuals with BEN frequently require dialysis and may ultimately succumb to kidney failure or may develop urothelial cancer. The accumulated

[2] Thyroid peroxidase is the enzyme responsible for catalyzing the reactions involved in the formation of the thyroid hormones triiodothyronine and thyroxine. Inhibition of thyroid hormone synthesis or release from the thyroid gland is involved in goiter formation.

evidence indicates that BEN is an environmentally induced disease and may affect 25,000 individuals with more than 100,000 at risk (U.S. Geological Survey 2001). Several potential causal agents have been hypothesized and investigated, including lead, cadmium, selenium deficiency, aristolochic acid, and mycotoxins (Stefanovi 1999). At present, it is believed that the most likely cause of the disease is exposure to groundwater contaminated by water-soluble organics derived from Pliocene lignites (Stefanovi 1999).

The disease occurs only in rural villages located on alluvial valleys of tributaries to the lower Danube River (Feder et al. 1991). A common geologic feature of endemic villages is the proximity to distinctive low rank Pliocene lignite deposits and lignitic shales. It is hypothesized(Feder et al. 1991) that weathering of the lignites and associated shales yielded soluble organic compounds that are transported by the local groundwater flow system to the shallow water wells used by the villagers. Geochemical analysis of water samples from endemic and control villages (Feder et al. 1991) indicates the presence of potentially nephrotoxic organic compounds (including napthylamines, aniline, aminophenols, and PAHs) in much higher concentrations compared to non-endemic villages. Many of these compounds are known to be carcinogenic, and may be the cause of urinary tract tumors that frequently accompany BEN in Bulgaria and Yugoslavia (Petkovi 1968; Chernozemsky 1977). Exposure to organics from lignite- and peat-associated water has also been reported to be associated with the high incidence of urothelial cancer in Alaska, Maine, Minnesota, North Dakota, and South Dakota (Tatu et al. 1998).

## Multiple Sclerosis

Multiple sclerosis (MS) is one of the most common diseases of the central nervous system. Excluding trauma, MS is the most frequent cause of neurological disability in early to middle adulthood and is estimated to affect 250,000 to 350,000 people in the U.S. (Hauser 1994). It is characterized by chronic inflammation, demyelination, and scarring of the central nervous system. During the disease process, the myelin sheath, which surrounds and insulates nerves, is destroyed leaving behind plaques or lesions that appear as hardened scars. MS is generally believed to be an immune-mediated disease that occurs in genetically susceptible individuals (Noseworthy et al. 2000). MS is not usually a fatal disease; however, it frequently results in severe disability and decreased quality of life (Sadovnick 1997).

Although the etiology of MS is unknown, both genetic and environmental theories have been proposed. Most researchers agree that MS is a disease with multifactorial etiology, and that both genetic susceptibility and environmental influences are important determinants in the acquisition and clinical expression of MS (Noseworthy et al. 2000; Hogancamp et al. 1997; Marrie et al. 2000; Kahana 2000). MS has been reported to be associated with exposure to soil and/or water high in organic content, such as that resulting from the leaching of coal deposits. Investigators reported that MS was significantly associated with exposure to groundwater in the Canadian province of Saskatchewan, an area which is located in the global northern band known for a high prevalence of MS (Irvine et al. 1993). The prevalence of MS in this province has been reported to be greater than one patient per thousand individuals (Irvine et al 1993). Geological studies of this region have identified areas of kerogen-rich White Speckled Shale that may contribute to an increased concentration of organic compounds in the groundwater (Dunn and Irvine 1993). Scientists investigating the MS cluster

hypothesized that consumption of this groundwater may have been linked to the cluster. In addition, Finnish researchers observed a correlation between the distribution of acidic soils, particularly the presence of acid raised bogs in Europe, and an increased prevalence of MS (Häsänen et al. 1986).

# CONCLUSIONS

The formation of coal from woody material produces a complex chemical matrix rich in aromatic structures. Thermal stress and oxidation reactions can cleave chemical bridges linking these aromatic structures, resulting in the production of water-soluble oxygen-, nitrogen- and sulfur-bearing water-soluble organics. The abundance of such water-soluble organics produced from these processes is related to coal rank and composition. Less mature coals will have the potential to yield greater amounts of water-soluble organics than more mature coals. The suite of water-soluble organic compounds that have been identified in coal-associated groundwater is that expected from a consideration of coal structure.

Two human diseases, goiter and BEN, have been associated with the consumption of groundwater derived from coal, likely due to the presence of organic compounds. Potent goitrogens have been detected in groundwater derived from coals, and their presence in water supplies could pose a serious threat of thyroid disorders. It has also been reported that ingestion of dissolved organics present in coal-associated water may produce a serious wasting disease of the kidneys, may induce urinary tract tumors, may increase the incidence of digestive tract cancers, and may be associated with the occurrence of MS clusters.

In an effort to put CBM-produced water to a beneficial use, Pennaco Energy is currently reinjecting CBM water into an aquifer used by the city of Gillette (Tollefson 2000). However, based on the possible suite of dissolved organics that may be present in this water, and the potential for human consumption, this method of handling CBM water should be carefully reviewed. Likewise, dissolved organics present in CBM-produced water may also be harmful to fish and wildlife. Consequently, the environmental impact of surface discharge of CBM water should be evaluated to determine the ecotoxicological impact of dissolved organic compounds. In conclusion, the injection of CBM-produced water into drinking water aquifers should not be undertaken until the organic constituency of such water has been thoroughly characterized and reviewed for potential risks to human and ecological health.

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**Table 1.** Water production in some major CBM-producing basins (modified from Rice 2000).

| Basin | State | Number of CBM Wells | Av. Water Production (bbl/day/well) | Water/Gas Ratio Bbl/Mcf | Primary Disposal Method |
|---|---|---|---|---|---|
| Black Warrior | AL | 2,917 | 58 | 0.55 | Surface Discharge |
| Powder River | WY, MT | 2,737 | 400 | 2.75 | Surface Discharge |
| Raton | CO | 459 | 266 | 1.34 | Injection |
| San Juan | CO, NM | 3,089 | 25 | 0.031 | Injection |
| Uinta | UT | 393 | 215 | 0.42 | Injection |

**Table 2.** Median and range for carbon, oxygen, hydrogen, and nitrogen content and gross appearance of coals of various ranks[3].

| Rank | C (%) | O (%) | H (%) | N (%) | Appearance |
|---|---|---|---|---|---|
| Lignite | 7369-76 | 21<br>16-25 | 5.1<br>4.5-6.0 | 1.2<br>0.1-1.75 | Many plant fragments still identifiable |
| Sub-bituminous | 76<br>67-84 | 17.4<br>11-26 | 5.4<br>3.5-7.1 | 1.3<br>0.1-2.3 | Cell structure of plants largely degraded |
| Bituminous | 84<br>71-92 | 8.5<br>1.2-22 | 5.6<br>3.6-7.1 | 1.6<br>0.1-2.6 | Black, hard and bright; breaks into rectangular lumps |
| Anthracite | 93<br>91-97 | 1.8<br>0.4-4.2 | 3.5<br>1.2-4.2 | 0.9<br>0.6-2.1 | Black, shiny rock with conchoidal fracture |

[3] Numerical data compiled from the World Wide Web subset of the Penn State Coal Sample Bank and Database available at http://www.ems.psu.edu/COPL/coal.html, downloaded August 22, 2002; values for carbon, oxygen, hydrogen and nitrogen are dry, mineral matter free weight %.

**Table 3.** Median and range of carbon, oxygen, hydrogen, nitrogen, sulfur, and volatile matter content of Wyodak-Anderson coal.[4]

| C (%) | O (%) | H (%) | N (%) | S (%) | Volatile Matter |
|---|---|---|---|---|---|
| 75.76 | 17.9 | 5.33 | 1.11 | 1.16 | 46.02 |
| 73.11 – 76.18 | 16.63-21.03 | 4.65-6.17 | 1.02-1.37 | 0.43-1.85 | 44.86-54.74 |

[4] Numerical data compiled from the World Wide Web subset of the Penn State Coal Sample Bank and Database available at http://www.ems.psu.edu/COPL/coal.html, downloaded August 22, 2002; values for carbon, oxygen, hydrogen and nitrogen are dry, mineral matter free weight %; sulfur is total, dry weight basis.

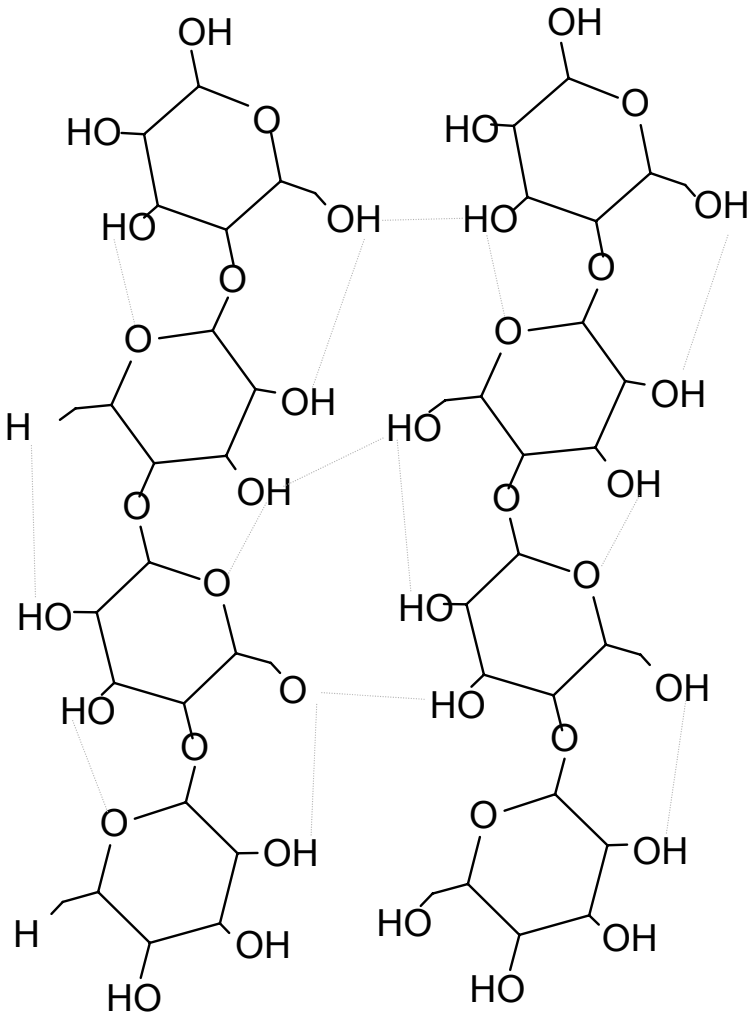


**Figure 1.** Structure of cellulose.

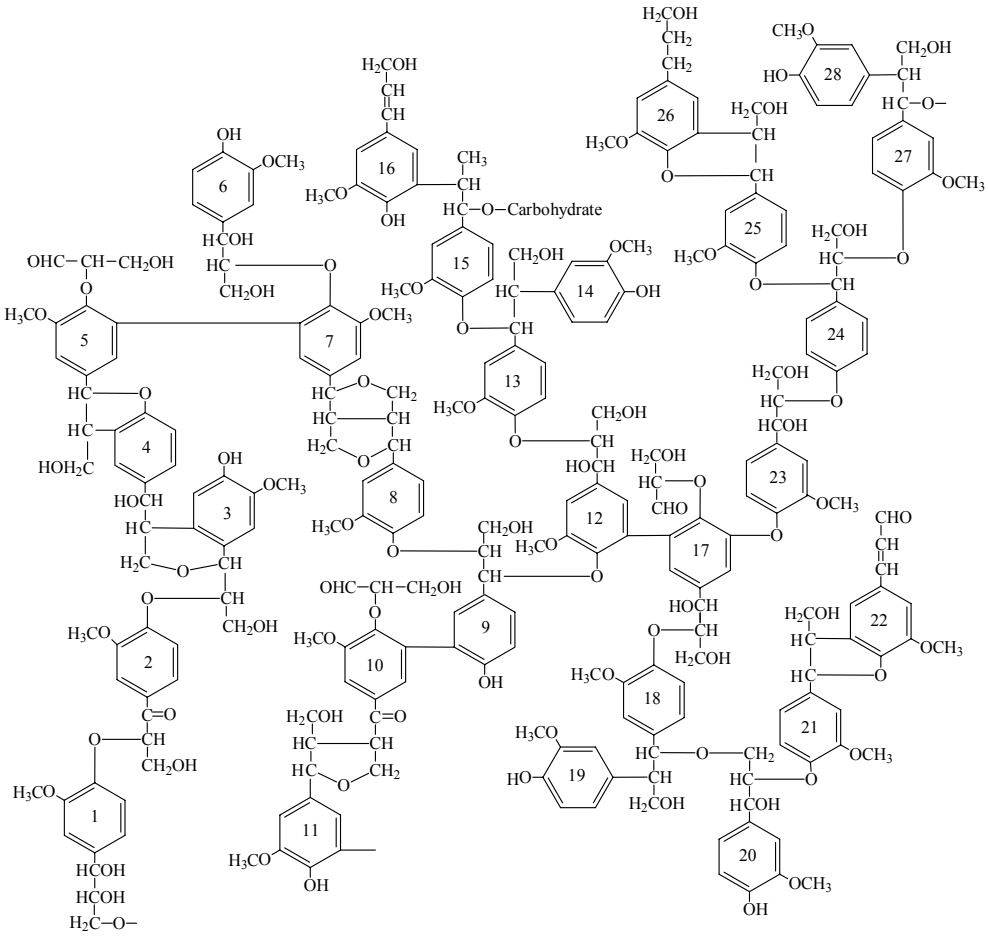


**Figure 2.** Proposed softwood lignin structure (Sakakibara 1991).

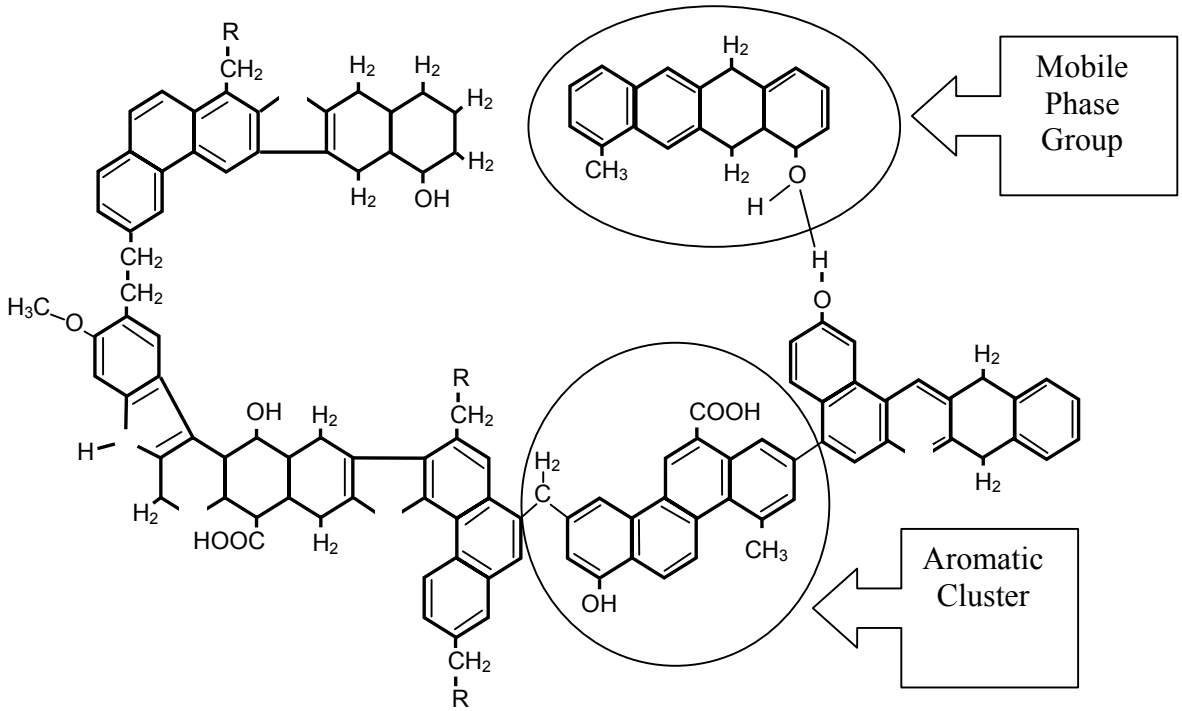


**Figure 3.** Schematic Chemical Structure of Bituminous Coal Showing an Aromatic Cluster and a Mobile Phase Group (modified from Solomon et al. 1988).