

Methane gas: An unconventional energy resource

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Methane gas in the form of 'coal bed methane' and 'hydrate', probably the last remaining hydrocarbon, is waiting to be exploited as an alternative source of energy. Coal plays the role of source rock as well as reservoir for coal bed methane. Hydrate is a unique chemical compound of methane and water found in deeper sections of ocean floor sediments.

UNITED States and Canada have been exploring methane obtained from coal beds ('coal bed methane') since the early 1970s and 1980s, respectively. Production of coal bed methane for domestic energy needs has grown significantly only in USA. Other coal-producing countries like Australia, China, Russia, Germany, Great Britain, Poland, etc. including India too have paid attention to the exploration of this new resource (now being considered as an economically viable unconventional source of energy) and have initiated several research programmes on different aspects of coal bed methane.

In fact, irrespective of their maturity (rank), organic composition, or nature of occurrence, all coals contain gases, with methane usually as the main constituent (90–97%). However, deep-seated seams (depth around 1000 m or more) containing coal of higher rank (carbon content > 83.5%, R_0 max 0.7% and above) are considered most suitable for commercial extraction of methane. The global coal bed methane reserves are presumed to be several times greater than the total reserves of all the known conventional gas fields.

Around 1969, gas hydrate deposits were first discovered in Russia (Siberian gas fields). As a result, interest in methane from sea floor ('hydrate') began. Natural occurrence of methane hydrate was noticed in other regions of oceanic and terrestrial environments by the 1970s. The encouraging results from discoveries at Blake Ridge (USA) opened up new vistas to consider gas hydrates as a potential source of energy for the future. Currently, countries like Mexico, Japan and India have launched national projects for the exploration of methane hydrate, the most abundant carbon fuel resource.

Conditions of extreme high pressures and cold temperatures favour accumulation of methane hydrates in deep ocean floor sediments, especially along continental margins. It is believed that methane hydrate reserves could possibly hold more fossil fuel energy than is present in conventional oil, gas and coal deposits.

Coal bed methane

Methane gas is generated during the formation of coal through 'coalification' process of vegetal matter (Figure 1). This can broadly be divided into biochemical and physico-chemical stages of coalification incorporating five successive steps¹:

Peatification (anaerobic degradation of organic materials in the peat swamp);

Humification (formation of dark coloured humic substances by anaerobic degradation);

Bituminization (generation of hydrocarbons with increase in temperature and pressure);

Debituminization (thermal degradation of matter and generated hydrocarbons); and

Graphitization (formation of graphite).

An excellent summary of the coalification process is given by Levine². Many physical and chemical changes, governed by biological and geological factors, occur during these processes. Whereas darkening in colour and increase in hardness and compactness are the main physical changes, loss in moisture and volatile contents, and increase in carbon content are the main chemical changes. Many acids (humic, fatty, tannin, gallic, etc.) and dry and wet gases (CH_4 , CO_2 , N_2 , N_2O , H_2S , ethane, propane, butane, etc.) are formed during decomposition of the organic matter. All the changes brought about are attributable to the release of $-\text{COOH}$ (carboxyl), $>\text{C}=\text{O}$ (carbonyl), $-\text{OH}$ (hydroxyl) and $-\text{OCH}_3$ (methoxyl) functional groups from the organic compounds which cause the decomposition of vegetal source matter.

Biochemical stage of coalification, beginning with the accumulation of vegetal matter and terminating at the sub-bituminous stage of coal formation, leads to the formation of a wide range of degradational products – the organo-petrographic entities of coal (termed 'macerals') by the partial oxidation and hydrolytic decomposition of dead vegetal matter accumulated in water-saturated wet lands (basins/grabens) by micro-organisms (fungi, aerobic bacteria, insects, etc.). Further decomposition by anaerobic bacteria extracts oxygen

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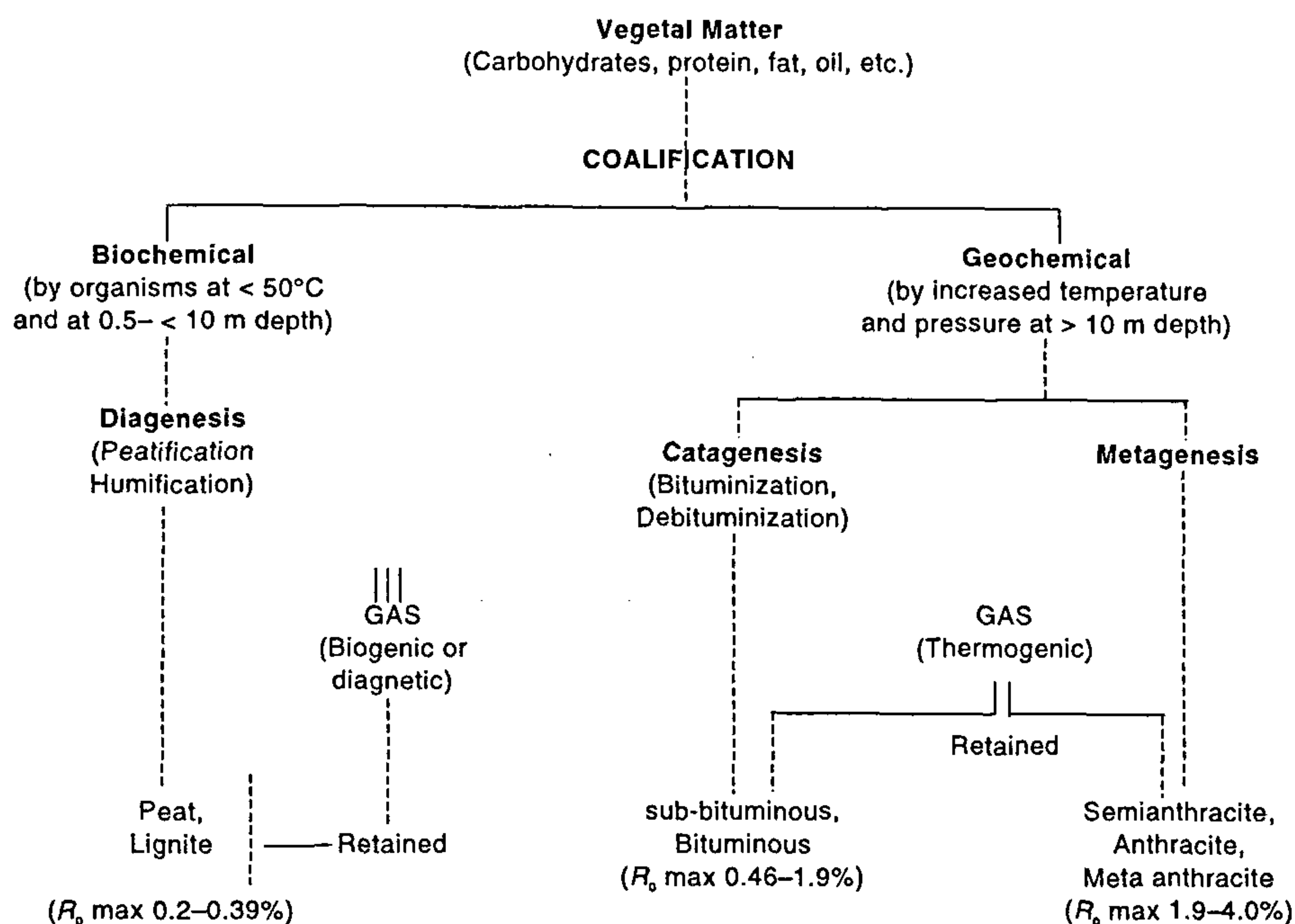


Figure 1. Flow-chart of methane gas generation during coalification.

from organic molecules of vegetal matter and results in high concentration of hydrogen. Part of this hydrogen is released as methane or 'marsh' gas and the rest is absorbed by humic colloids.

During subsequent geochemical stage of coalification, rising temperatures and pressures, due to subsidence of the basin/graben, either by growing thickness of overburden or by tectonic activities, generate hydrocarbons (hydrogen-rich constituents). Thermal cracking of the free lipid hydrocarbon fraction and/or cracking of the kerogen fraction of coal generates methane gas. Thus, the generation of coal bed methane during coal formation occurs in two ways:

- (i) by metabolic activities of biological agencies (biological process), and
- (ii) by thermal cracking of hydrogen-rich substances (thermogenic process).

Methane generated at shallow depths (<10 m) and lower-rank stage (sub-bituminous) by the first process (active up to 50°–80°C) is termed 'biogenic' or 'diagenetic methane' (Figure 1). Methane generated during this process is about 10% of the total methane generated by subsequent steps of coalification (catagenetic: > 80°–150°C, R_0 max > 0.50–2.0% and metagenetic: > 150°–200°C, R_0 max > 2.0–4.0%).

Though most of the gas generated during early stages of coalification generally escapes into the atmosphere through the exposed peat or due to low hydrostatic pressure, some amount can accumulate under certain specific geologic conditions like rapid subsidence and burial, and thus may get trapped in shallow reservoirs.

Gas produced at greater depths and higher rank stages of the second process, the thermogenic methane, constitutes bulk of the coal bed methane. The gas generation, by this process, begins at vitrinite reflectance (R_0 max) values of 0.70–0.80%, peaks near the boundary between medium-volatile bituminous and low-volatile bituminous coal stages [R_0 max 1.1–1.4% (maximum at 1.2%), temperature 100°–150°C (ref. 3), Figure 2 a], and declines further with the rise in temperature and reflectance values^{4,5}. Thus, it could reasonably be presumed that the prospect of generation of coal bed methane is more in the regions of high palaeogeothermal gradient as well as in the vicinity of intrusive bodies.

Although, methane is the major gas component of coal gases; water, carbon dioxide, wet gases and liquid hydrocarbons are also released during coalification. Total amount of methane generated during the coal formation (between R_0 max 0.5–2.0%) approximately ranges between 2000 and >5000 Scf/ton (ref. 6) (Figure 2 b). However, part of methane generated is retained in coal beds/seams and is termed 'coal bed methane' (CBM);

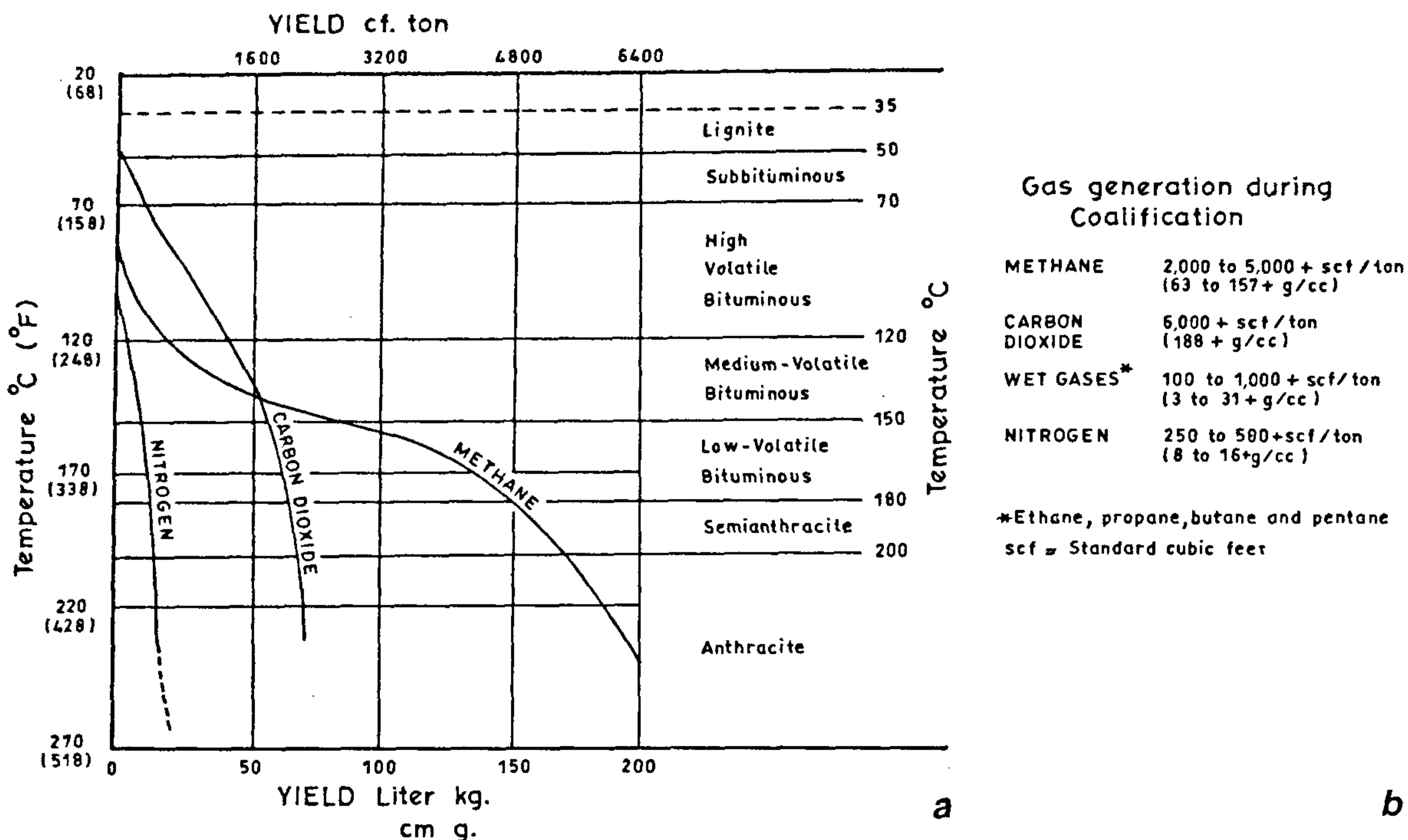


Figure 2. a, Relationship between gas generation and coal rank (source Manjrekar³); b, Gas volumes generated during coalification up to vitrinite reflectance values of 2.0% (after Scott⁶).

and the excess above the retention capacity of the coal bed, tends to migrate to the surrounding reservoir rocks (e.g. sandstones). Retention of methane in the coal beds is⁷:

- as 'adsorbed' molecules on internal surfaces or 'absorbed' within the molecular structure of the coal
- as gas molecules held within the matrix porosity (macro- and micro-porosity)
- as free gas within the fracture network
- as gas dissolved in groundwater within the coal bed.

Since methane is generated during coal formation processes, all coals invariably contain methane. However, the gas content of the coal normally increases with (i) rank of the coal, (ii) depth of burial of the coal seams, provided the roof and overburden are impervious to methane and (iii) the thickness of the coal seams.

Content of coal bed methane is assessed by several factors, amongst which the rank of the coal is the most important. According to Tang *et al.*⁴ economically important quantities of methane (> 300 Scf/ton) are generated by thermogenic process, since large quantities of gas production are impossible until a certain threshold

of thermal maturation is attained. This requirement is met in the high-volatile A bituminous rank at R_0 max between 0.8 and 1.0%. Investigations, world over, have shown that high rank coals buried at great depths (> 300–1200 m or more) are suitable for coal bed methane exploration, provided certain other geological and inherited coal seam characteristics are favourable as well.

Generally, gas is more concentrated in geologically active areas, such as folded and faulted regions as well as the surrounding areas of the faults. The well-developed cracks and fractures in the coal seams owing to tectonic disturbances, provide permeability to coal seams. The permeability of the seam is also related to the cleat system present in coals. The sealing capability and thickness of the seam roof and floor rocks play a significant role in methane accumulation. The fluvial basins, having higher rate of subsidence accompanied by thermal events and moderate tectonism, are the prospective sites for the exploration of coal bed methane.

The methane-generating capacity of coal is, however, related to the coal macerals. Whereas macerals of the vitrinite and liptinite (or exinite) groups are the greatest contributors of methane gas, the macerals of the iner-

inite group have relatively little hydrocarbon generating potential, though they have the greatest capacity for storage of methane. Of the exinite groups, the liptinite macerals have the highest gas generating potential. Besides the cleats and other fracture systems, the mesopore structure of certain macerals (including structured inertinite) significantly enhance the permeability of methane within the coal seams. Therefore, vitrinite-rich coals of higher rank are reasonably the most important sources of coal bed methane for they have more micro-porosity (that is much higher absorbing capacity) than the other two maceral groups. Ash content of the coal also has an influence on the coal bed methane content: lower the ash content; higher is the gas content of the coal seam.

Methane gas sorbed on coal particles can be liberated by desorption of coal seams. The gas pressure in coal seams is released either by dewatering the coal seams or by drilling borewells which facilitate the flow of gas through fractures. As stated earlier, the amount of methane produced depends on desorption capacity of coals, which varies from coal to coal depending on its physical and chemical properties, especially the type of coal (*sensu* maceral composition). The amount liberated however, may be enhanced by using stimulation techniques as have been practised in USA. Existing techniques of methane production being expensive, many companies are engaged in developing appropriate technologies for cost-effective production of this gas.

Status of coal bed methane in India

The prospect for coal bed methane is mainly related to the coal resources of the country. India has huge Gondwana (mainly Permian, 99.5%) and Tertiary (Eocene and Oligocene) coal deposits distributed in several basins located in peninsular and extra-peninsular regions. About 204 billion tons of coal reserves have been established and approximately 200 million tons or so are likely to be added in the near future by further explorations. The main Gondwana coal basins are rifted intra-cratonic grabens having thick sequence of coal seams, and hold considerable prospects for coal bed methane. The major part of Indian Gondwana coals (mostly up to 300 m depth) is of low rank, far below the threshold value of thermogenic methane generation. However, high rank coals, amenable for generation of coal bed methane, mostly occur in untapped deeper parts of basins covered by younger sediments.

Tertiary coals of India, occurring mainly in lagoonal to deltaic sediments, are better in quality compared to Gondwana coals, though the seams are thinner. On the basis of composition and rank of coal, Tertiary coals appear to be moderately rich in coal bed methane. The estimated coal bed methane resource of Gondwana coals

appears to be between 1 and 1.5 Tcm and the Tertiary coals of about 4.3 Bcm (ref. 7).

In 1990, efforts to exploit coal bed methane were initiated by Essar Oil (a private oil company) under the advice of American experts. The methane emission and desorption studies on Gondwana coal samples from Jharia Coalfield (Bihar) were carried out by Central Mine Planning and Design Institute Limited (Ranchi) and Central Mining Research Institute (Dhanbad). The content of gas and gas emission rate from these samples were found to be 1.8–2.3 m³/1000 m² of surface and 12.7–17.3 m³/min, respectively⁸. The studies carried out by Bharat Coking Coal Limited in the same area with the help of French experts indicated 0.68–1.45 m³/min gas emission rate.

In 1992, assessment of coal bed methane potential for Damodar Valley coals⁷ was initiated by Oil and Natural Gas Commission. Till date, it has collected significant data related to coal bed methane exploration from drills in Raniganj basin. Recently, ONGC for the first time in country has succeeded in flowing the gas from seam no. XIV in Parbatpur block of Jharia basin⁹.

Besides, Geological Survey of India and Reliance Industries Limited have also undertaken investigations on the prospects of occurrence of coal bed methane in different Gondwana and Tertiary coalfields of India. These investigations led to the delineation of potential areas in Damodar (Raniganj, Jharia, Bokaro, Giridih), Son (Sohagpur) and Pench-Kanhan-Tawa Valley (areas lying on the dipside of the Kanhan Valley) coalfields where a total gas-in-place reserve of 13.34 Tcf has been predicted¹⁰. In addition, gas content of 250 Scf/ton in an area of 900 km² has been recorded from Early Paleocene coal beds (50 m thick) of Cauvery Basin by Essar Oil Company¹¹.

Methane hydrate

Presence of methane gas in coal seams has been known since the very beginning of the coal researches. However, its presence in ocean bottom sediments as 'hydrate' is a relatively recent discovery. Mysterious disappearance of ships, sudden plane crashes and many strange events on land have unfolded the mystery of 'Bermuda Triangle' in Atlantic Ocean (*The Times of India*, Lucknow edition, 11 January 1998, published through *The Sunday Times*, London). The Bermuda Triangle, earlier considered as the area of supernatural power, is now known as a powerful source of energy with huge accumulation of methane gas in the form of hydrate. According to United States Geological Survey, two very small areas of north and south Carolina coast lying in a part of Bermuda Triangle contain gas equivalent to about 70 times the annual gas consumption of USA. The encouraging findings point towards high po-

tentiality of methane hydrate as an alternative source of energy. Energy density of methane in hydrate is about 10 times greater than the non-conventional sources of gas and 2–5 times greater than that of conventional natural gas.

In 1810, hydrate (of chlorine) was first discovered by Humphry Davy, when he noticed the formation of ice-like crystals upon cooling of an aqueous solution¹². In 1934, an American chemist, E. G. Hammerschmidt, first identified the hydrate of methane in frozen ice that was obstructing the flow of gas in natural gas pipes in Russia¹³.

Hydrates are naturally occurring solids of a special category called the 'inclusion compounds' in which a molecule of chemical component gets trapped within a molecule of another component without any chemical bonds between them. 'Clathrate' is one of the inclusion compounds in which the molecule to be caged-in fits into spherical voids of another molecule and can be released only by the decomposition of the compound. If the host molecule is water it is called 'clathrate hydrate' or 'hydrate', and if the guest molecule is a gas then the term 'gas hydrate' is used.

Thus, gas hydrates are the ice-like compounds or solids formed by physical combination of a natural gas (mainly methane) and water, under specific conditions, in which water crystallizes in the isometric crystallographic system rather than the hexagonal system of normal ice. All gases except hydrogen, helium, and neon form gas hydrates. Presence of sufficient amounts of gas and water under conditions of extreme cold (temperature $<7^{\circ}\text{C}$) and high pressure (> 50 atmospheres) are the prerequisites for hydrate formation, restricting thereby the natural occurrence of methane gas hydrates to deep sea sediments (~30–2000 m beneath sea floor, 300–500 m below sea level) and permafrost zones, on the outer continental margins and shelves¹⁴.

Various reflection techniques have been adopted to locate gas (methane) hydrate zones. It was first suspected to exist at the sea floor on the Blake-Bahama Outer Ridge, off the coast of USA¹⁵. Several reports on the presence of gas hydrates at permafrost zones and bottom sediments have been published since then^{16–20}. Global surveys using bottom simulating reflector (BSR) have shown the presence of hydrate zone along the continental sea slopes almost all along the world²¹.

Several regions showing high concentrations of gas hydrate have been identified in the offshore and onshore permafrost regions between New Jersey and Georgia and Alaska, in USA; and in Siberia and Norway. Intensive explorations carried out by oceanographers and oil geologists have also brought to light several sites of gas (methane) hydrates. Presence of canyons, which carry organic matter, at the continental rise provide supporting evidence for a hydrate zone. Thus, methane hydrates

occur in polar regions, shallow offshore sediments in cold regions (permafrost), and deep sea sediments in tropical regions. Presently, occurrence of approximately 2×10^4 Tcm methane as methane hydrate is estimated globally.

Excessive methane gas which gets trapped inside the water molecule to form hydrate, is mainly biogenic. However, contribution of thermogenic gas in hydrate formation cannot be ruled out. In fact, deeply buried fine organic matter along with sediments has the potential to generate thermogenic gas, at elevated temperatures (80° – 150°C), that migrates upwards and is trapped in the hydrate zone below the 'blanket' of hydrate²¹. Biogenic methane is produced at low temperatures by the action of micro-organisms on rapidly buried organic matter at the ocean bottom sediments (Figure 3). For the production of methane²², sedimentation rate $> 30 \text{ m Ma}^{-1}$, an organic carbon content exceeding 0.5%, and residual methane content $> 10 \text{ ml l}^{-1}$ are required. Evidently, the most significant factor controlling methane hydrate accumulation appears to be the rate of sedimentation. Areas of rapid hemipelagic times are favourable to accumulate considerable amounts of organic detritus and preserve them from oxidation by rapid burial at the sea floor so that it is converted into abundant methane by the bacteria within the sediments²³.

Methanogenesis, i.e. methane formation only begins on the onset of complete anoxic conditions as the methane-producing bacteria are strict anaerobes and are inactive even in the presence of traces of oxygen. In marine environments, methane in excess is generated by carbonate reduction in a complete sulphate-deficient condition²⁴ (Figure 3). Methane generated in large amounts by the reduction of carbon dioxide, remains as solution in interstitial pore water because of its high solubility at high hydrostatic pressure caused by the weight of the overlying water column. It exudes as free gas from interstitial pore water either on supersaturation or by the release of hydrostatic pressure.

Methane thus generated, forms icy-white crystals of methane hydrate at different water depths under favourable temperature and pressure conditions. The hydrate formed in hydrate stability zone (HSZ) forms a hard 'blanket' of impermeable hydrate. This 'blanket' traps gas migrating from deeper levels. Depending on temperature–pressure conditions, hydrates can form at a depth of 335 m and 610 m in Arctic and sub-tropical waters, respectively²⁵ (Figure 4). The thickness, lower limit and stability of the hydrate zone, are controlled by geothermal gradient which increases with depth (varying locally) as we proceed vertically downwards across the hydrate zone. Due to increase in temperature at greater depths hydrate becomes unstable, dissociates, and accumulates as free gas.

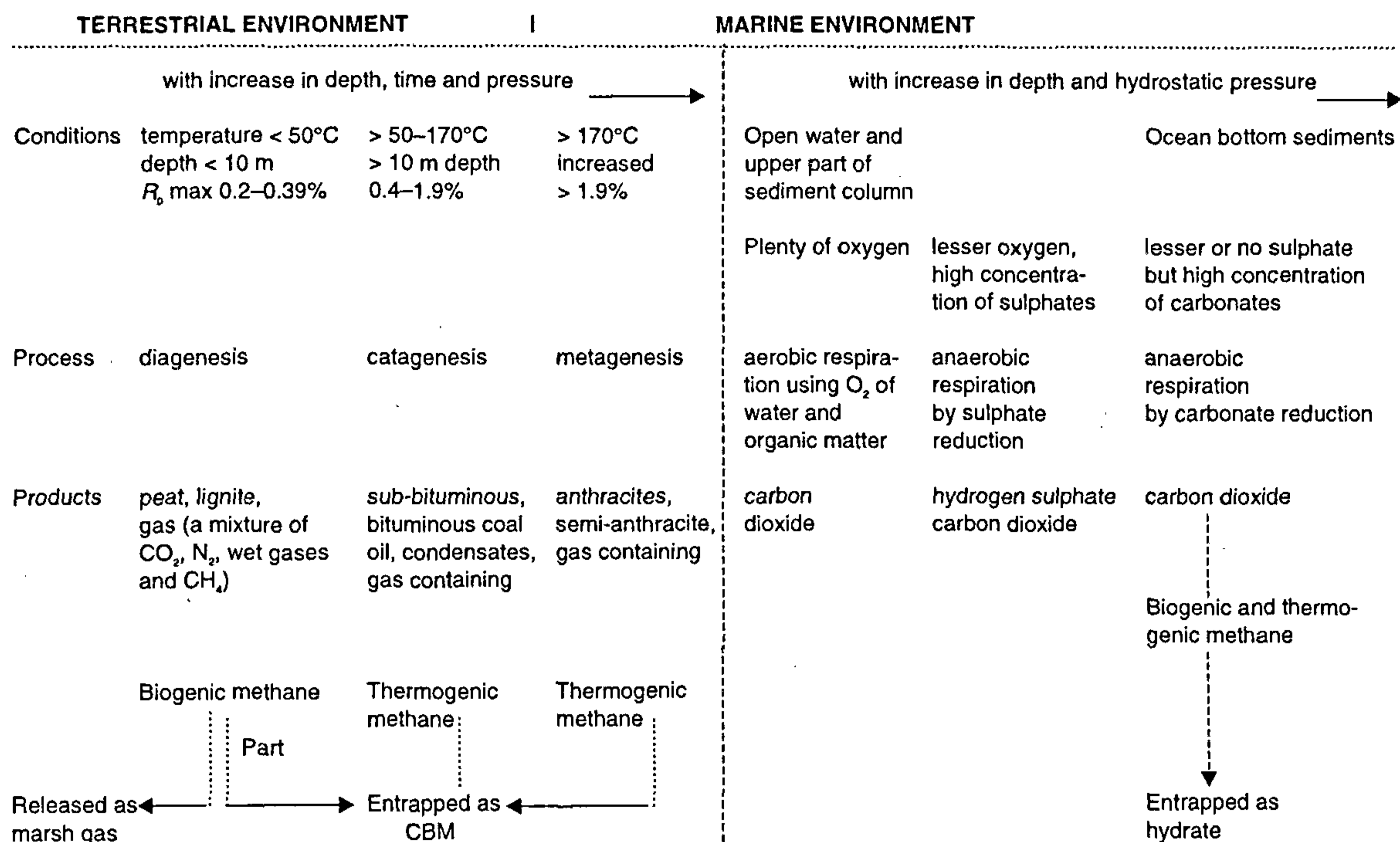


Figure 3. Schematic representation of methane generation in terrestrial and marine environments.

In the oceanic sediments, gas hydrates are stable when the ocean bottom-water temperature approaches 0°C , and the water depth exceeds 300 m. On the other hand, hydrates expand at normal temperature and pressure, and release large amounts of gas locked up inside the ice crystals. It is estimated that 1 m^3 of fully saturated hydrate releases approximately 167 m^3 of methane gas, and a puddle of water, on expansion.

Discovered since the 1970s, hydrates are still in embryonic stage of research and development. Gas hydrate exploration and exploitation is currently a frontier area of research all over the world. Hydrated methane can be recovered by the following ways: dissociation of hydrate crystal either by increasing temperature in hydrate-bearing zone (thermal stimulation method) or by reducing pressure (depressurization method). Inhibitor injection method disturbs the temperature–pressure equilibrium and releases methane from hydrate. An appropriate econo-technology for production of methane from hydrate is yet to be developed.

Status of methane hydrate in India

At present only the resource estimation and delineation of prospective areas for methane hydrate has been done.

Preliminary study of seismic attributes from Indian deep water offshore by Oil and Natural Gas Commission has shown the presence of large reservoirs of methane hydrate at depths of 1000 to 3000 m, and has suggested that Krishna–Godavari and Andaman–Nicobar Islands may be explored for hydrates. About 7.5 Tcm of methane is estimated in an area of about $80,000\text{ km}^2$ from Indian deep offshores, which is about 5 times the total conventional gas reserves of the country⁹. Besides, Gas Authority of India Limited is carrying extensive investigations on methane hydrates.

Indian continental margins (especially on the east coast in Bay of Bengal) with excess sedimentation rate and organic carbon content than required for methane hydrate production are the potential sites for methane hydrate exploration²⁶. The physical parameters (temperature, pressure, salinity) controlling the formation of methane hydrate are also met at the site at a water depth ranging from 650 m (east coast) to 750 m (west coast)^{9,27}.

Conclusions

Methane (CH_4), the second most significant greenhouse gas after carbon dioxide (CO_2) is mainly responsible for

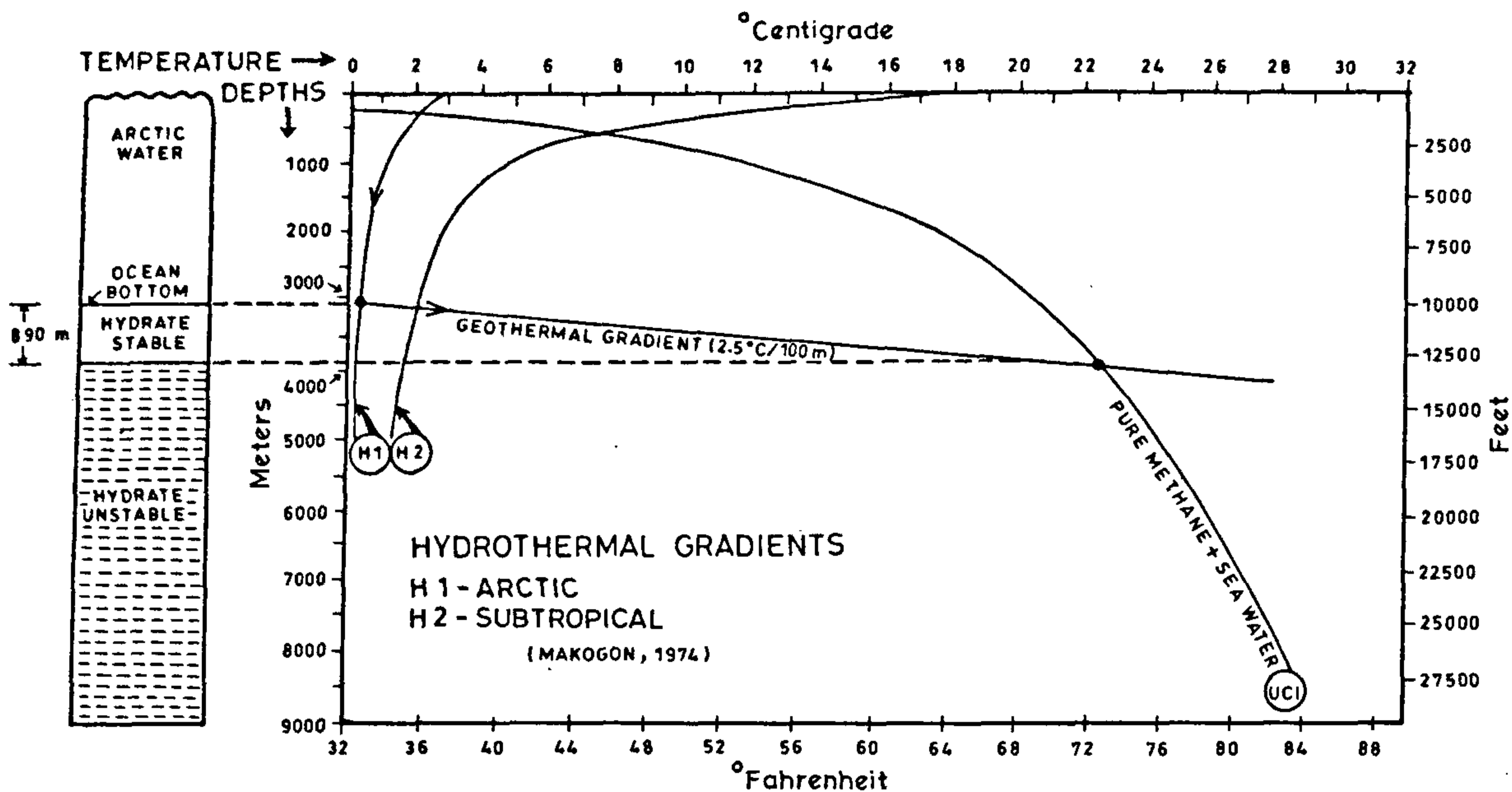


Figure 4. Temperature–depth (pressure) phase diagram of gas hydrate stability (after Macleod²⁵).

ozone holes and global warming; traps atmospheric heat approximately 24-times more than carbon dioxide.

Thus, conversion of the most hazardous gas methane into a commercial energy resource, besides solving the world's energy problem for the coming century, will be advantageous in many other ways as mentioned below. The hazards involved in the exploitation of the hydrate deposits require development of suitable methods. Consumption of methane, which has the highest heat retaining capacity and the lowest atmospheric life-time span (about 10 years), will mitigate the global warming at a much faster rate. Consequently, methane hydrate – our future powerhouse – adversely affected by global warming, will be saved.

Emergence of methane gas as an additional energy resource warrants rational utilization and preservation of coal deposits, which are both the 'producer' and 'reservoir' of this gas and besides, burning can also be utilized for the exploration of methane gas. These vast energy reserves can be saved by selective utilization of coal.

Many leading nations have already launched projects to extract methane gas from coal beds and hydrates. However, continuous release of methane gas into the atmosphere during degasification of coal mines, by chlorofluoro carbon (cfc) compounds, and by hydrates adversely affects the global climate as well as human beings. Furthermore, the highly explosive nature of this

gas, resulting in serious fires and accidents on land, water and air, has raised a few concerns on its utilization as a source of energy. At present, India too has all the scientific and engineering skills required for the production of methane from coal bed and seabeds. There are substantial unconventional energy (CBM and CH₄-hydrate) resources, but their exploration and economic exploitation from inaccessible areas are great challenges.

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REVIEW ARTICLE

Lipoprotein (a): Biology and role in atherosclerotic vascular diseases

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Lipoprotein (a) (Lp(a)) is a genetically determined lipoprotein molecule. Its constituent, low density lipoprotein cholesterol participates in the process of atherosclerosis, and its prothrombotic tendency is due to its homology to plasminogen. In normal subjects plasma levels of Lp(a) are controlled by the apolipoprotein(a) (apo(a)) gene locus. The polymorphism of apo(a) is determined by more than 34 alleles, plasma levels of Lp(a) being inversely correlated to its isoforms. Plasma levels in healthy subjects are highly variable, and also depend on the ethnic group studied. Indians, both immigrant and native, display high plasma level of Lp(a). Many studies record them to have higher levels than most of the other ethnic groups. It is now established as a

powerful and independent risk factor for macrovascular diseases due to atherosclerosis, including coronary artery disease (CAD), stroke and peripheral artery diseases. High levels of Lp(a) (> 30 mg/dl) appear to increase the risk of premature CAD. The risk is increased several-fold in the presence of high levels of other lipid and non-lipid risk factors. Lp(a) plasma concentrations are abnormal in other diseases, nephrotic syndrome and chronic renal failure and possibly in insulin-dependent diabetes mellitus as well. Unfortunately, this lipoprotein is not readily amenable to therapeutic intervention. Predictive value of this lipoprotein, and its observed high plasma levels makes it an important research, investigative, and prognostic tool, particularly for the Asian Indian population.

FACTOR(S) presumed to be responsible for increased propensity to atherosclerosis in Indians are abdominal obesity, smoking, glucose intolerance, resistance to the insulin-mediated glucose uptake, and a peculiar dyslipidemic profile^{1,2}. The dyslipidemia seen in Indians include high

triglycerides (TG), low high density lipoprotein cholesterol (HDL-c), and increased lipoprotein(a) (Lp(a)). A new dimension has been provided by the emerging role of Lp(a) and its importance in atherosclerosis-prone Indians. The following discussion shall outline in detail the genetics, biochemistry, metabolism, and clinical relevance of this lipoprotein.

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