

CARBON SEQUESTRATION IN COAL SEAMS IN JAPAN AND BIOGEOCHEMICAL CARBON CYCLE IN TERTIARY SEDIMENTARY BASINS

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ABSTRACT

A new research and development project “CO₂ sequestration in coal seams” began in Japan in 2002 to develop new type of “coal mines” for CO₂-enhanced coal gas recovery. Coal seams are promising for CO₂ storage and as sources of hydrocarbon gas. Researches are being devoted to a deeper understanding of CO₂-coal interaction, to monitoring of CO₂ behavior in coal seams, to prospecting of untapped deep coal seams, etc. Supercritical CO₂-enhanced coal seam gas recovery (ECGR) and in situ fire-free microbial gasification of coal can produce sufficient amounts of methane for the CO₂ emission-free closed-circuit power plant and new type of “coal mines” without underground works, even in gas-poor coalfields. Underground biogeochemical carbon recycling makes zero-emission closed-circuit power and heat generation possible, while coal seams and saline aquifers are capacious enough to accommodate large volume of CO₂ from fossil fuel and refuse-derived fuel (RDF).

Tertiary sedimentary basins are suitable as CO₂ stores, hydrocarbon gas reservoirs and possibly CO₂-CH₄ converters. Subsurface biogeochemical carbon recycling can realize greenhouse gas control with restoration of energy resources.

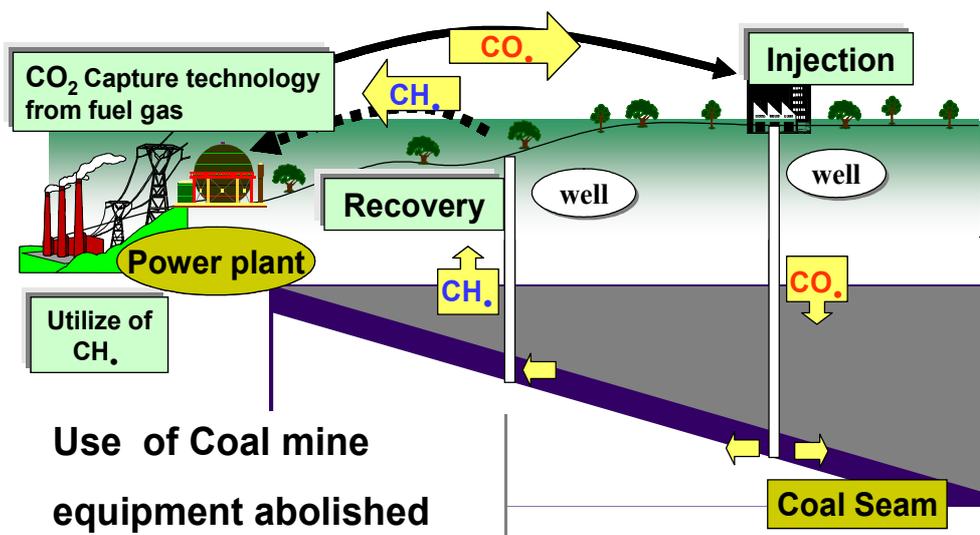


Figure 1: CO₂ emission-free closed-circuit system of electric power generation by CO₂-ECGR (enhanced coal seam gas recovery)

CO₂ SEQUESTRATION IN COAL SEAMS

The Taiheiyou Coal Mine in Hokkaido, northern Japan was closed on January 30, 2002, although a new company “Kushiro Mine” took over the operation of the last underground coal mine in Japan at a reduced scale.

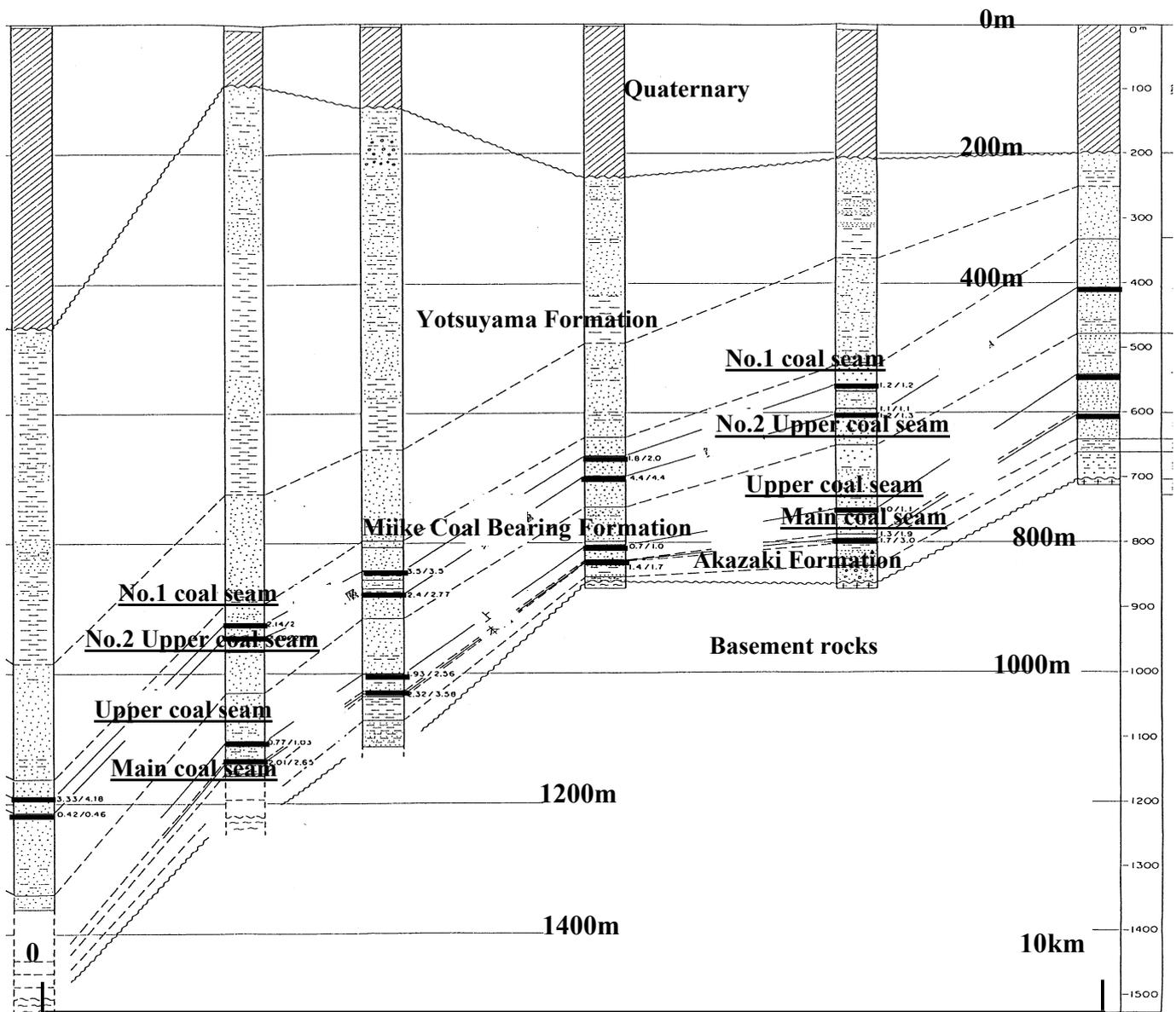


Figure 2: Geological section of the Ariake Coalfield

Deep untapped coal seams as well as remaining coal in old mines provide huge potential CO₂ reservoirs and sources of hydrocarbon gas. CO₂ sequestration into coal seams with recovery of coalbed methane can realize the CO₂ emission-free closed-circuit system of electric power generation [2]. The last two major mines-Taiheiyou mine and Ikeshima mine- were undersea mines. However, offshore extensions of many coal seams were out of survey. Coal seams deeper than 1200m are excluded from the official coal reserve in Japan. Unminable coal seams deeper than 1200m and offshore coal seams provide possible reservoirs for CO₂ sequestration and potential targets for enhanced coal seam gas recovery (ECGR). The 18.2 billion tons of technologically minable coal reserves in Japan may contain 254 billion m³ of coal seam methane. Technologically minable coal seams in Japan can adsorb almost 1 Gton of CO₂ because the coal can roughly adsorb CO₂ twice as much as methane [3]. Yamaguchi and Yamazaki [4] estimated that about 2.5 trillion cubic meters of methane are contained in unminable coal seams in and around Japan under the assumption that unminable coal is some 10 times of known coal reserves. Unminable coal seams may adsorb almost 10 Gton of CO₂ in and around the Japanese Islands. New type of “coal mines” have large potential without underground labors but need technological, geological, economical and environmental investigations with special respect for safety. Japan Coal Energy Center (JCOAL), other 19 business members and 18 individual members founded Japan Forum on CO₂ Sequestration in Coal Seam in 2001. The forum started a research on CO₂ sequestration in coal seams and coal seam gas recovery. In 2002, the Agency for Natural Resources and Energy (ANRE) in the Ministry of Economy, Trade and Industry (METI) launched a new research and development project “CO₂ sequestration in coal seams”(Table 1). METI provided 246 million yen for the first year of the first phase of new national project. Kansai Environmental Engineering Center

Co., Ltd.(KANSO) and JCOAL are main players of the R&D project including preliminary field tests on CO₂ sequestration in coal seams and coal seam gas recovery in a coal mine and on CO₂-ECGR in deep virgin coal seams. Research Institute of Innovative Technology for the Earth (RITE), Non Profit Organization (NPO) Think-tank Kyoto Institute of Natural History, Hokkaido University, Akita University, Kyoto University and Waseda University contribute to the fundamental researches of the national project. Kansai Electric Power Co. Inc., Mitsubishi Heavy Industries, Ltd. etc are also among the R&D team. The fundamental researches include studies on CO₂-CH₄ replacement mechanism, CO₂ behavior simulation, the optimum condition for CO₂ sequestration, CO₂ adsorption mechanism and CO₂ seepage behavior in coal seams. The development of CO₂ injection-CH₄ recovery technology, suitable CO₂ capture technology from flue gas, system analysis for operation, and investigation of CO₂ sequestration capacity in inland and offshore coal seams in and around Japan are also among main subjects of the R&D project. Monitoring technology for CO₂ sequestration in coal seams (Table 2), environmental impact assessment, safety assessment and public outreach are emphasized in the geologically unstable Japanese archipelago.

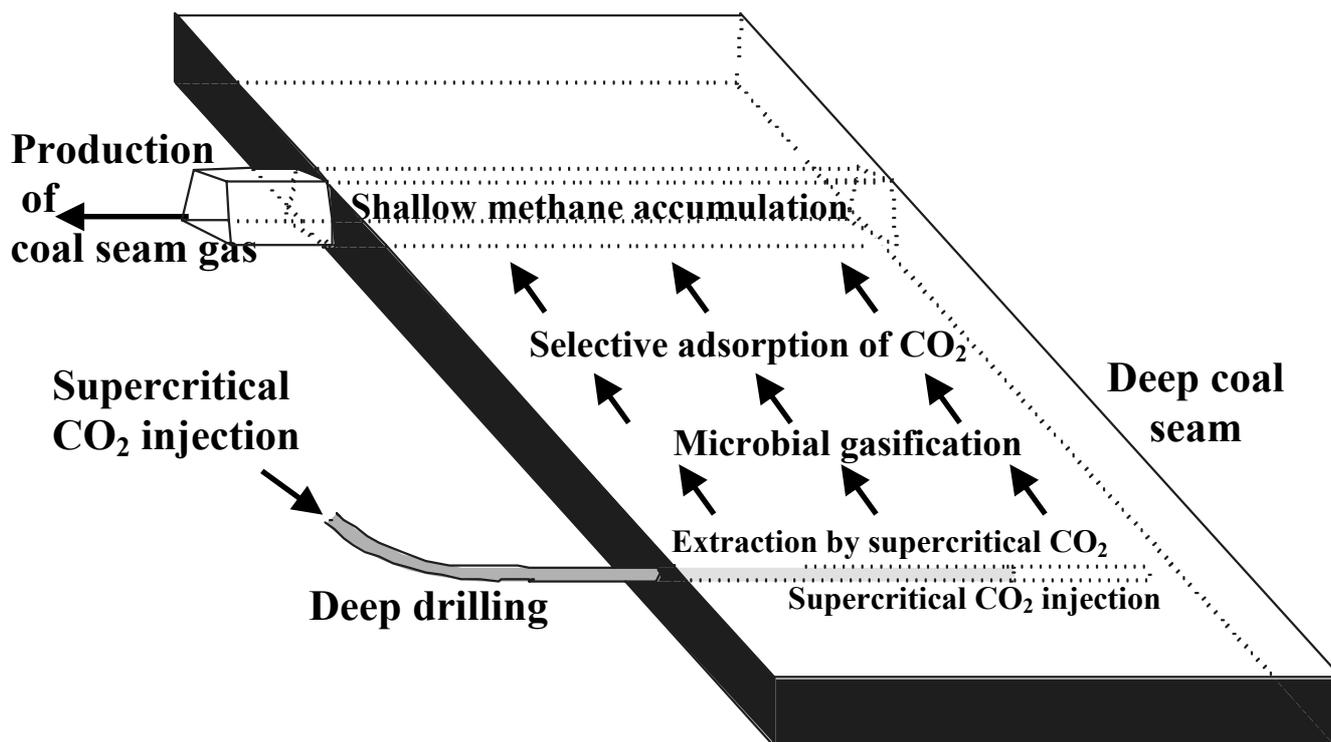


Figure 3: Supercritical CO₂-enhanced coal seam gas recovery (ECGR) and in situ fire-free microbial gasification of coal

SUPERCritical CO₂-ECGR AND IN SITU MICROBIAL GASIFICATION OF COAL

The authors made a preliminary study on the possibility of CO₂ sequestration in off-shore unminable coal seams with gas recovery in Japan. The first study was made on the Ariake Coalfield under the shallow Ariake inland sea. The Ariake Coalfield is the offshore extension of the old Miike Coal Mine that was the largest coal mine in Japan with the maximum annual coal output of 6.57 million tons in 1970 but closed in 1997. Offshore extensions of main four coal seams- Main, Upper, No.2 Upper and No.1 coal seams- in the old Miike Mine become deeper in the westward-dipping Paleogene Miike Coal Bearing Formation in the Ariake Coalfield (Fig.2). Measured adsorption capacity of the Miike coal is 72.3 kg-CO₂/ton-coal on an average. Estimated CO₂ adsorption capacity of the coal seams in the Ariake coalfield is 8 million tons in the Main coal seam, 8 million tons in the Upper coal seam, 9 million tons in the No.2 Upper coal seam and 7 million tons in the No.1 coal seam. Hence, total CO₂ sequestration capacity in the submarine Ariake coal seams is about 32 million tons-CO₂. CO₂ is highly soluble in water under high pressure. CO₂ sequestration is also possible in dissolved state in groundwater in pores of underground rocks. Paleogene rocks in the Ariake coalfield are abundant in sandstone of average porosity 7.04%. Average geothermal gradient is 3.2°C/100m. Pore fluid pressure and temperature can be estimated from the depth. CO₂ solubility is estimated from fluid pressure and temperature. The Miike Coal Measure can sequester 104 million tons of

CO₂ and upper Paleogene sandstone of the Yotsuyama Formation can sequester 174 million tons of CO₂ in dissolved state in groundwater. The Paleogene formations in the Ariake coalfield are covered by Quaternary impermeable clay layers. Therefore, coal seams and Paleogene sandstone in the submarine Ariake Coalfield can accommodate total 300 million tons of CO₂ in dissolved state in groundwater.

The Miike coal contains only 6 m³-methane/ton-coals on an average [5]. Coal seam methane is too poor to realize closed-circuit power generation in the Ariake-Miike Coalfield. However, almost pure CO₂ can extract volatile components of coal as a supercritical fluid at high pressure. The critical pressure of CO₂ is 7.39 MPa. The critical temperature of CO₂ is 31.1°C. Almost pure CO₂ (99.9%) is extracted from the flue gas using monoethanolamine (MEA) or improved solvent KS-1[6]. Injected pure CO₂ behaves readily as a supercritical fluid around injection wells in deep coal seams (Fig.3). Pressured CO₂ dissolve in groundwater around supercritical fluid zone. Microbial gasification is ready in the mixture of coal, CO₂, water and extracted carbon compounds. Finally, selective adsorption of CO₂ and desorption of methane in coal enrich the hydrocarbon gas mixture (Fig.3). In the laboratory, 35 to 50% of coal can be converted to methane using genetically adapted bacterial cultures, which is equivalent to 534 to 763 m³/ton [7]. Supercritical CO₂-enhanced coal seam gas recovery (ECGR) and in situ fire-free microbial gasification of coal can produce sufficient amount of methane for the CO₂ emission-free closed-circuit power plant and new type of “coal mine” without underground works even in gas-poor coalfields.

BIOGEOCHEMICAL CARBON CYCLE IN TERTIARY SEDIMENTARY BASINS

In Japan, Paleogene sedimentary formations often contain bituminous coal seams but Neogene sedimentary basins contain only lignite, peat, coaly materials or other fossil organic matter. However, lignite, peat, coaly materials and fossil organic materials in black shale can be sources of biogenic methane. Sedimentary basins often contain abundant organic materials. Subsurface microbes combined with effects of compaction, tectonic load and geothermal heat decompose buried organic materials and produce abundant natural gas. Methane-rich gas dissolved saline groundwater and adsorbed in coal seams suggests active methanogenesis in sedimentary basins.

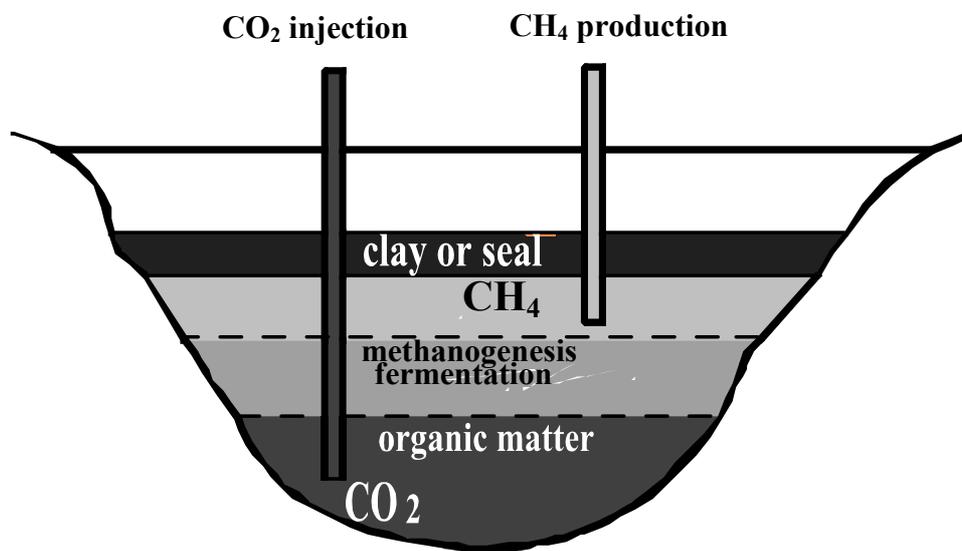


Figure 4: Microbial carbon recycling in a sedimentary basin

Chemolithotrophic microbes can fix CO₂ into organic acids. Eventually methanogens reduce CO₂ into methane. Coal seams, coaly matters or tuffaceous layers adsorb CO₂ to prevent migration. Saline aquifers in deep sedimentary basins dissolve CO₂ to prevent leakage. Injection of CO₂ into deep coal seams or saline aquifers induces entrapment of CO₂ and exsolution of methane as methane is ready to migrate upward than CO₂. Counter extraction of methane from shallower formations prevents emanation of methane into the atmosphere and supplies a low-carbon fuel (Figure 4). Methanogenesis will restore biogenic gas in the sedimentary basin. Underground biogeochemical carbon recycling makes zero-emission closed-circuit power and heat generation possible, while coal seams and saline aquifers are capacious enough to accommodate large volume of CO₂ from fossil fuel and refuse-derived fuel (RDF). Tertiary sedimentary basins are suitable as CO₂ storages, hydrocarbon gas reservoirs and possible CO₂-CH₄ converter. For an example, around Lake Biwa, Shiga Prefecture, Southwest Japan, methane gas has been used for source of energy in domestic usages since more than one hundred years ago. The methane gases are impregnated in sedimentary layers under high pressure in subsurface reducing environment. The methanogens are strong candidate to generate

methane from carbonic materials and carbon dioxide in anoxic sediments. An underground closed-circuit “methane factory” is proposed to process waste gas of a bio-power plant in the bottom of sedimentary basin where the CO₂-rich waste gas and organic materials are converted into CH₄-rich biogenic gas. Preliminary survey of the southwest part of Kanto sedimentary basin, too, revealed that late Pliocene- early Pleistocene saline sandstone aquifer can store about two billion tons of CO₂ in the dissolved state. The southwest Kanto aquifer is covered by mudstone layers and close to major CO₂ sources in the Yokohama area.

RITE and NPO Think-tank Kyoto Institute of Natural History started a new research and development program on “underground microbial CO₂ sequestration and methane factory (2002-2004)”. RITE also started another new R&D program on “hydrate and geochemical sequestration of CO₂ in deep underground and ocean bottom environments (2002-2004)” for advanced stable carbon isolation [8, 9]. Subsurface geochemical and biogeochemical carbon recycling may realize greenhouse gas control with restoration of energy resources (Figure 5). Underground anaerobic microbial carbon recycling provides a new access to hidden another natural carbon cycle driven by geothermal energy and untapped fossil energy without solar energy. Monitoring and analysis of carbon cycle in geosphere and study of geosphere-biosphere interaction are needed for greenhouse gas mitigation and comparable solution of energy problems.

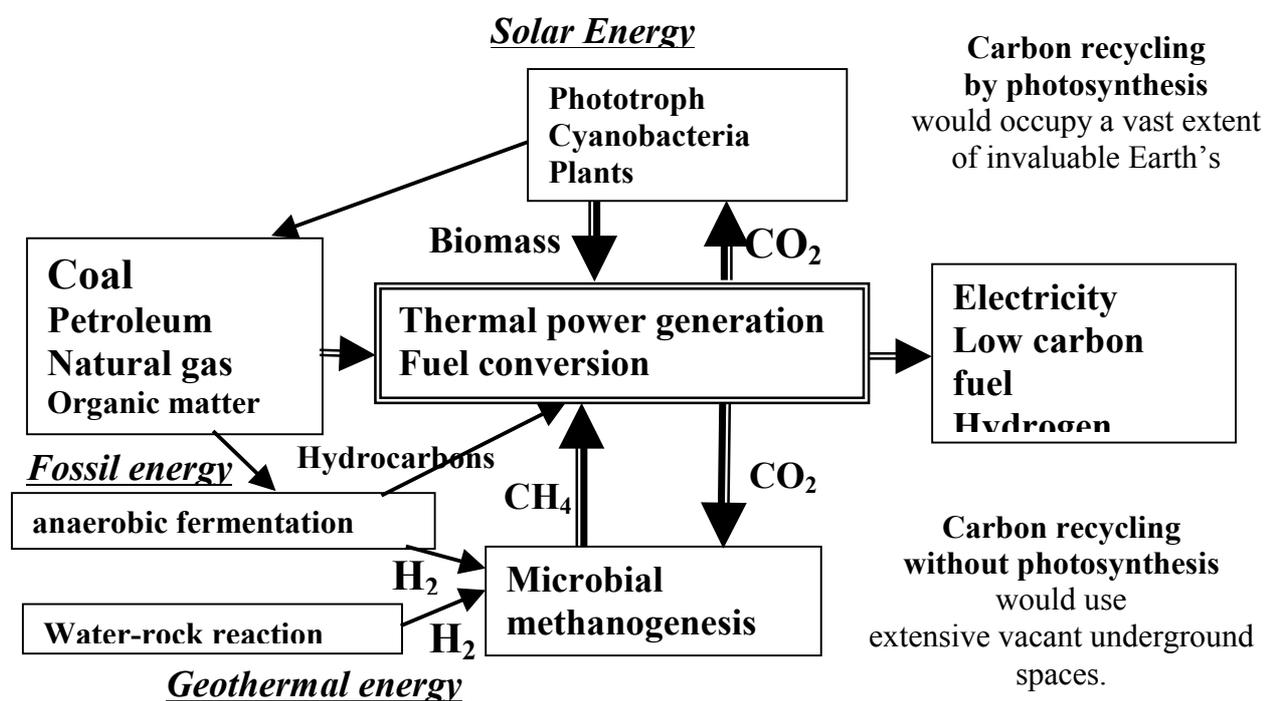


Figure 5: Underground anaerobic microbial carbon recycling

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