

AQUIFER DISPOSAL OF CO₂ AND NATURAL GAS RESERVOIRS

Hitoshi KOIDE
Geological Survey of Japan
1-1-3, Higashi, Tsukuba, Ibaraki 305, Japan

Masaki IJIMA
Mitsubishi Heavy Industry Ltd.
15-1, Tomihisa, Shinjuku-Ku, Tokyo, Japan

Yoshiyuki TAZAKI
Kanto Natural Gas Development Co.
661, Mobara, Mobara, Chiba, Japan

Yoshikazu NOGUCHI
Electric Power Development Co.
6-15-1, Ginza, Chuo-Ku, Tokyo, Japan

Yuji SHINDO
National Institute of Materials and Chemical Research
1-1, Higashi, Tsukuba, Ibaraki 305, Japan

ABSTRACT

Some of natural gas reservoirs with high content of CO₂ suggest capability of long-term containment of CO₂ in the depth of sedimentary basins. Separation of CO₂ from methane and its disposal are inevitable to develop natural gas deposits with high content of CO₂ such as the Natuna natural gas deposit, Indonesia, natural gas of which contains about 71% of CO₂. There exist huge volumes of useless aquifers in sedimentary basins of the world due to high salinity of water. Small fractions of useless aquifers in terrestrial sedimentary basins in the world are enough to host the whole amount of carbon dioxide which is produced directly from extraction of natural gas and by burning of fossil fuels.

INTRODUCTION

Huge volumes of CO₂ are emitted into the air by burning of fossil fuels which have been extracted from underground reservoirs. The carbon dioxide is not very poisonous but may enhance the greenhouse effect to cause artificial global warming. However, the intensive use of fossil fuels as the energy source makes the development of effective technology for mitigation of man-made emission of carbon dioxide extremely difficult. About 60% of CO₂ emission in Japan are from fixed massive sources such as electric power plants, mills, etc. Intensive efforts are made in Japan for development of recovery technology of CO₂ from flue gas of power plants and mills. However, great questions remain on the space accommodating the enormous volumes of separated carbon dioxide. The ocean disposal of CO₂ is the most investigated but is still troubled with marine environmental problems.

The carbon contained in underground rocks is almost 100,000 times of atmospheric carbon. Fossil

fuels contain a small fraction of underground carbon, but still reach about 5.5 times of atmospheric carbon (Berner and Lasaga, 1989) which is enough to cause the significant greenhouse effect. It seems a natural solution to return the carbon dioxide into underground rocks after burning of fossil fuels.

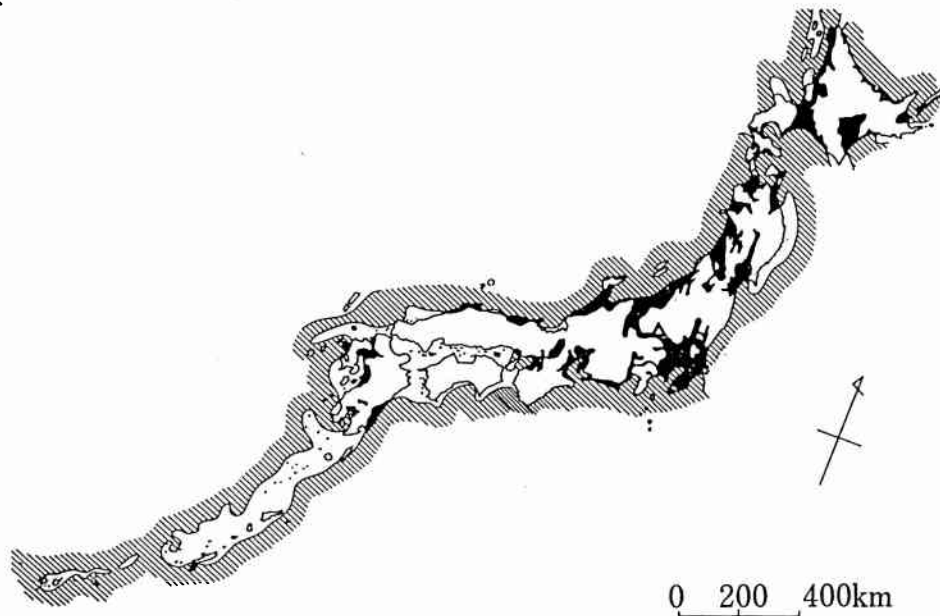


Fig.1 Location of the Isobe natural carbon dioxide field(Δ) and sedimentary basins(shaded) where deep aquifers contain fossil water with dissolved methane.

NATURAL CARBON DIOXIDE GAS RESERVOIRS

The underground carbon dioxide is mostly fixed in carbonate rocks but there exist some free carbon dioxide in gaseous or supercritical state. Huge amount of carbon dioxide are dissolved in the groundwater in the forms of CO_2 , H_2CO_3 , CO_3^{2-} or HCO_3^- . There exist many hot and cool springs of carbonated mineral water in Japan. Natural or artificial carbonated mineral water is people's favorite drink which is essentially harmless to health.

Some of natural gas reservoirs contain almost pure carbon dioxide gas. The Isobe gas deposit in Japan produced natural gas of nearly 99.5% carbon dioxide content up to 1981 (Fig.1). The subterranean carbon dioxide is derived from the decay of organic matter, oxidation of hydrocarbons, igneous emanation, metamorphism of carbonate and/or solution of limestone by groundwater (Farmer, 1965). In U.S.A., about 46 million tons of carbon dioxide are sent through several hundred kilometers of pipelines from natural carbon dioxide gas fields in the Rocky Mountains area to oil fields in the western Texas in order to use for the enhanced oil recovery (Bonder, 1992).

The natural gas in the Natuna gas field, Indonesia, contains about 71 % (in volume) of carbon dioxide (Fig.2). Separation of carbon dioxide from methane and aquifer disposal of carbon dioxide are inevitable for the development of natural gas of high carbon dioxide content such as the gas in the Natuna field. Thermodynamic calculation suggest that, although equilibrium gas composition in the deep subsurface is a strong function of reservoir mineralogy, the content of carbon dioxide tends to increase in comparison to the content of methane in very deep natural gas reservoirs (Barker and



Fig.2 Location of the Natuna gas field(Δ).

Takach, 1992). Separation and disposal of carbon dioxide would become very important for future extensive development of natural gas in very deep reservoirs.

STORAGE OF CARBON DIOXIDE IN DEPLETED NATURAL GAS RESERVOIRS

The natural gas and petroleum have been stored in underground reservoirs during millions of years. Impermeable cap rocks prevent the leakage of gas. Many depleted gas fields are used as temporary storage of natural gas. Depleted natural gas and oil fields are available for storage of carbon dioxide(Fig.3).

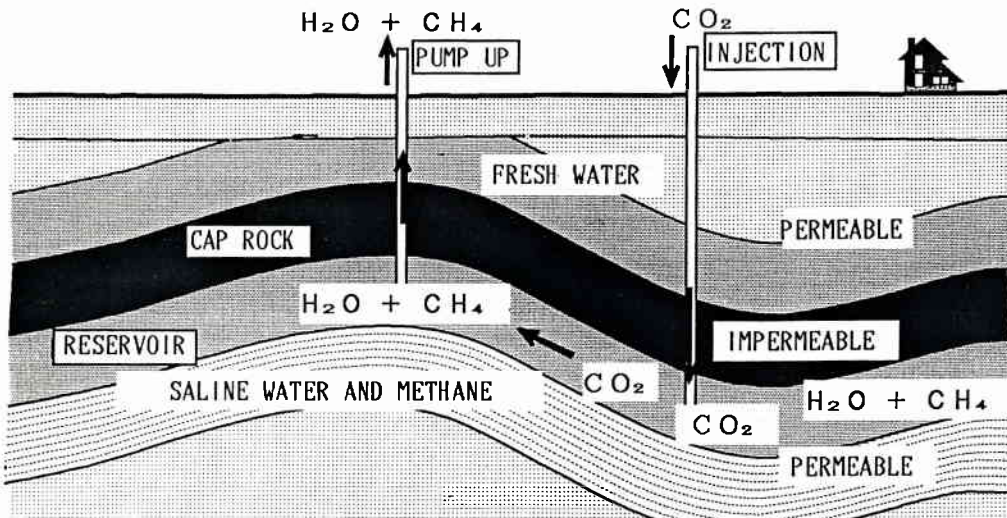


Fig.3 Injection of carbon dioxide into closed reservoir and extraction of methane

Deep high pressure reservoir can contain usually more amount of gas due to compression. It is expected that a depleted natural gas reservoir can safely contain gaseous carbon dioxide up to the primary formation pressure. A dry natural gas reservoir can theoretically accommodate same volume of gaseous carbon dioxide as extracted natural gas. On the other hand, burning of CH₄ produces almost same volume of carbon dioxide under the same pressure. Natural gas reservoirs of pure methane can store the almost same volume of carbon dioxide, at most, which is produced by burning of extracted natural gas.

Burning of 1 cubic meters of petroleum produces about 2.2 cubic meters of carbon dioxide in liquid. Therefore, oil reservoirs can store less than one half of carbon dioxide that is produced by burning of extracted petroleum. Natural gas and oil deposits cannot provide real sink of carbon dioxide.

AQUIFER DISPOSAL OF CARBON DIOXIDE

As the circulation of groundwater is very slow in rocks, deep aquifers tend to contain old groundwater which is not suitable for drinking nor for agricultural use due to high salinity. Impermeable layers trap very old fossil water of high salinity which is often the remain of sea water since the time of sedimentation of rocks. In oil and natural gas fields, huge volume of brine is trapped under oil and gas. There distribute widely useless aquifers in the depth of sedimentary basins in the world. Oil and gas exploration drilling rarely hits economically valuable deposits but often encounter useless aquifers which contain saline groundwater. The sea water intrudes under the sea floor and often wedges into deep inland aquifers. Flow of underground sea water is expected to be extremely slow.

Saline aquifers provide suitable reservoirs for disposal of carbon dioxide(Fig.4), because saline water is useless as ordinary water resources, and because the groundwater flow in saline aquifer is extremely slow or restricted by impermeable layers. The presence of old fossil water provides the good evidence of the reservoir capability for long-term containment of dissolved carbon dioxide. Low electric resistivity of saline water is convenient to locate possible underground reservoir of carbon dioxide by electric prospecting such as magnetotelluric method(Takakura et.al.,1992).

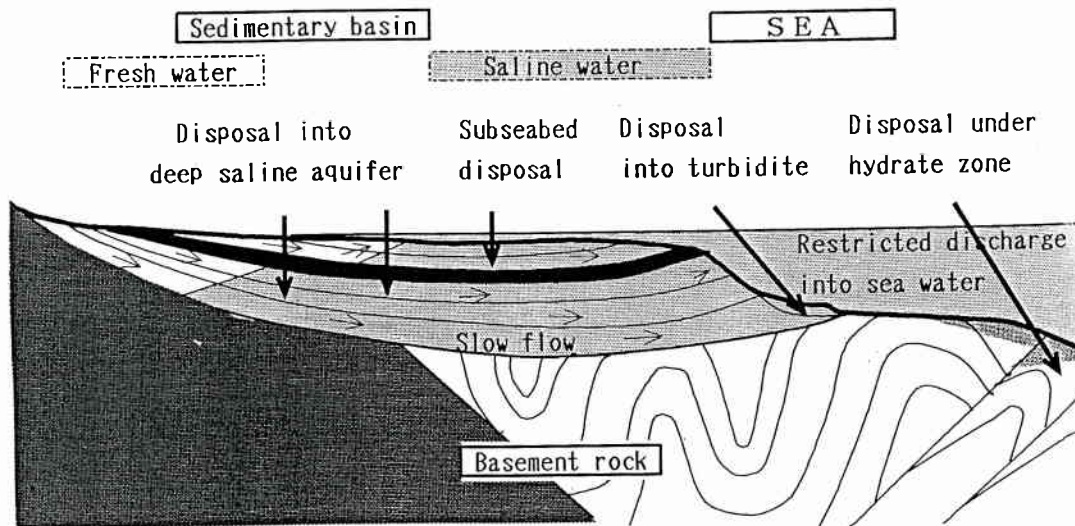


Fig.4 Sequestering of carbon dioxide into saline groundwater.

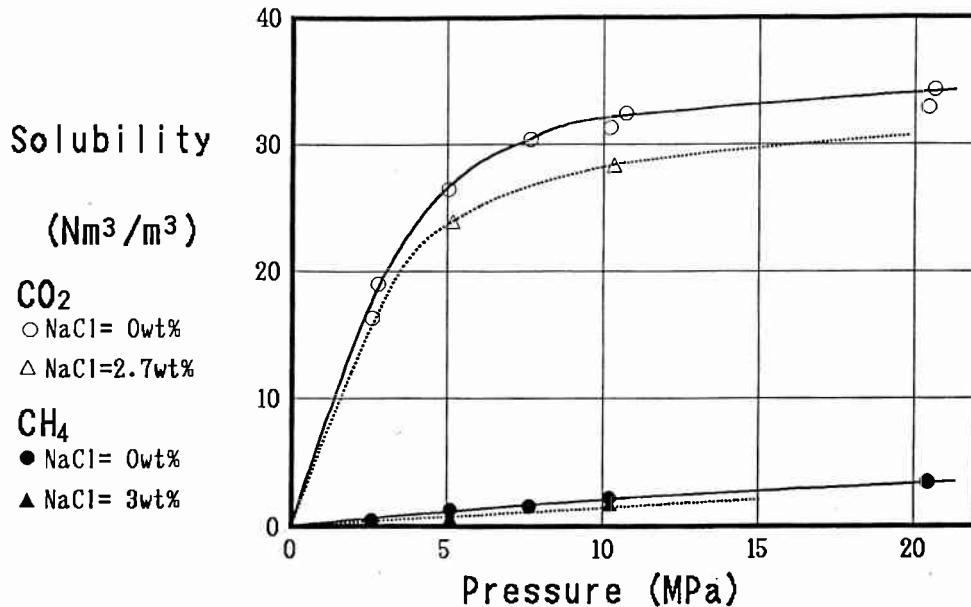
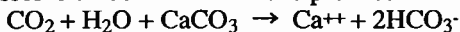


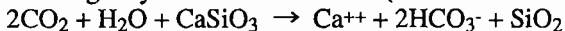
Fig.5 Solubility of carbon dioxide in pure water and in saline water at 30°C.

Fossil groundwater often contains hydrocarbons such as methane. In some regions of Japan, they pump up fossil groundwater, and extract the dissolved methane(Fig.1). Injection of carbon dioxide combined with pumping up of groundwater makes possible to recover the dissolved methane which exists extensively but vainly in fossil water in many deep sedimentary basins in the world(Fig.3). Carbon dioxide is much more soluble in water than methane under same temperature and pressure(Fig.5). We can extract methane and dissolve carbon dioxide as much as 20-40 times of methane instead into groundwater.

Carbon dioxide in groundwater forms carbonic acid. Although the carbonic acid is weak acid, concentrated solution of carbon dioxide under high pressure is highly acidic. Acidic groundwater dissolve carbonate minerals to produce bicarbonate ions.



The carbonic acid attacks silicate minerals, too. Highly simplified chemical formula of silicate rock weathering may be written as follows(Berner and Lasaga, 1989).



These processes of weathering of carbonate minerals and silicate minerals form bicarbonate ions to help containment of carbon dioxide and neutralize the groundwater.

Although mineralogy affects the chemical properties, groundwater is usually weak alkaline. When the solution of bicarbonate and calcium ions become alkaline, calcium carbonate would precipitate.



Precipitation of calcium carbonate may seal fractures to prevent further leakage of carbon dioxide. However, more study is necessary to clarify the behavior of carbon dioxide in various properties of groundwater and in various kinds of rocks.

The very conservative estimation that the suitable aquifers for CO₂ storage were to be found in depths of 1% area of sedimentary basins(Koide et.al., 1992) suggests that the CO₂ storage capacity of

sedimentary basins could reach well over 320 billion tons (=87gigaton-C) in the world.

The authors designed a prototype of CO₂ injection system into useless aquifers and estimated the storage cost(Koide et al.,1992). This system was assumed to inject 5,000 tons of carbon dioxide daily which was recovered from a fossil-fuel power plant. About 80 km² of aquifer will be necessary for storage of 5,000 tons of CO₂ per day over a period of 20 years. At an injection rate of 250 tons CO₂/day through a single well, 10 injection terminals, 20 injection wells and 30 pumping wells over an 8 km x 10 km injection site are necessary to inject 5,000 tons of CO₂ per day.

The total underground storing cost of CO₂ in aquifer is about 3,000yen/ton-CO₂, excluding the separation cost of CO₂ from flue gas. This does not include electricity charge, because recovered natural gas can compensate loss of electric power, although injection systems spend about 73kWh of electric energy per ton of carbon oxide.

Our preliminary cost estimation in Japan suggests that the CO₂-emission-free electricity generation may become possible with the cost increase of 35% for natural gas fired power station and of 60% for coal fired power station where carbon dioxide is separated from the flue gas by amines and injected into deep aquifers(Koide et al.,1993)

CONCLUSION

Separation of CO₂ from methane and its disposal are inevitable to develop natural gas deposits with high content of CO₂ such as the Natuna natural gas field,Indonesia. Saline aquifers provide suitable reservoirs for disposal of carbon dioxide, because saline water is useless as ordinary water resources, and because the groundwater flow in saline aquifer is extremely slow or restricted by impermeable layers. Useless aquifers of fossil water are found around oil and gas reservoirs and commonly in the depth of sedimentary basins. Small fractions of aquifers in terrestrial sedimentary basins in the world are enough to host about 320 gigaton of carbon dioxide.

A preliminary technical and economical survey on the carbon dioxide injection system suggests that the energy requirement for carbon dioxide injection into subterranean aquifers is about 73kWh /ton-CO₂ and that the investment and operation costs for the system are about 3,000yen/ton-CO₂. By our preliminary cost estimation in Japan, the CO₂-emission-free electricity generation may become possible with the cost increase of about 30-60%.

REFERENCES

- Barker, C. and Takach, N.E.,1992, Prediction of natural gas composition in ultradeep sandstone reservoirs, *The American Association of Petroleum Geologists Bulletin*, 76:1859-1873.
- Berner, R.A. and Lasaga, A.C., 1989, Modeling the geochemical carbon cycle, *Scientific American*, March:54-61.
- Bonder, P.L., 1992, Applications of carbon dioxide in enhanced oil recovery, *Energy Conservation and Management*, 33:579-586.
- Farmer,R.E.,1965, Genesis of subsurface carbon dioxide, *The American Association of Petroleum Geologists Memoir* 4:378-385.
- Koide, H., Tazaki,T., Noguchi,Y., Nakayama,S., Iijima,M., Ito,K. and Shindo,Y., 1992, Subterranean containment and long-term storage of carbon dioxide in unused aquifers and in depleted natural gas reservoirs, *Energy Conservation and Management*, 33:619-626.
- Koide, H., Tazaki,T., Noguchi,Y., Iijima,M., Ito,K. and Shindo,Y., 1993, Carbon dioxide injection into useless aquifers and recovery of natural gas dissolved in fossil water, *Energy Conservation and Management*, 34:921-924.
- Takakura,S.,Mitsuhata,Y.,Yokota,T. and Ogawa,Y., 1993, Integration of magnetotelluric and gravity data for resource assessment of Shimokita Peninsula, Report on Three-Dimensional Modelling Method for Resource Assessment, Geological Survey of Japan.