Thomas Gold
and the Future of Methane as a Fuel

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Abstract

Thomas Gold was a main participant and contributor in the controversy between the biogenic and abiogenic theories of the origin of hydrocarbons, a controversy launched by the abiogenic views of Mendeleev and supported by other Russian and Ukrainian authors. The great success of Gold's forecasts is illustrated by a photo of the methane seas on the cold planetary body Titan. Recently Scott et al.'s experiment on methane formation at high pressure suggests a possibility of methane formation in the mantle. Some thermodynamic equilibrium calculations suggest a possible exothermic reaction of carbon dioxide with fayalite producing methane. In this view, carbon could play the role of an energy carrier from fayalite to methane and then to a power plant and in a closed cycle be reinjected in Earth. Fayalite becomes a fuel, with methane the energy carrier. Methane is then a renewable energy source. The search for methane in Earth and resoluton of its origins deserve more efforts than ever before.

Introduction

The best known source of methane is natural gas. Methane can also be produced as biogas, typically from organic waste. In this case combustion of the fuel is carbon neutral, as it originates from photosynthesis from reduced CO_2. Recent evidence suggests that much more methane than previously suspected may be available to us. Many authors have discussed the availability of methane hydrates in which methane is "stored" inside a complex molecular structure. These hydrates could become an important source of methane in years to come. Deep gas pools also hold much unused methane. Expectations for exploration, development, and use depend upon the origin of hydrocarbons.

Biogenic theory states that hydrocarbons originated from debris of former organic life. They are located in the sedimentary layer, not too deep. Such views are popular up to now, especially in the West.

Abiogenic theory, first introduced by D. Mendeleev in 1877, predicts the production of hydrocarbon fuels by inorganic processes. N. Koudriavtsev greatly developed this theory (1,2). E. Tchekaliuk offered a thermodynamic background for oil formation (3). The best known recent advocate of abiogenic theory, Thomas Gold, discussed the production of hydrocarbon fuels simultaneously with other substances in the depths of Earth (4, 5). Gold's view led him to predict rivers and seas of methane on the surface of cold planets where organic life never existed.

Gold passed away in July 2004, 6 months before the European Space Agency's probe Huygens landed on Saturn's largest moon, the cold Titan (about –180° C), and made photos of the rivers and seas of methane Gold had predicted. As we see in Figure 1, shot from an altitude of 8 km, and Figures 2 and 3, made after landing 14 January 2005, the quantities of methane are astonishing. The distance from Titan to Earth was about 1.5 light-hours.

Origin of hydrocarbons

Starting from his essay "Rethinking the Origin of Oil and Gas" in the *Wall Street Journal*, June 8, 1977, Gold synthesized his popular and technical papers into two books (4,5). His central ideas might be expressed in a few sentences. According to Gold, our planet is like a sponge, filled by primordial hydrocarbons. They were formed together with the other substances of the deep Earth about 4 billion years ago. Due to their much lower density than the surrounding porous rocks, an upwelling of hydrocarbons, mainly methane, takes place from a depth of hundreds of kilometres. This permanent seepage from below fills the known pools of oil and gas, and some eventually escapes to the atmosphere. Hydrocarbons lose hydrogen along the way from initial CH_4 finally to $CH_{0.8}$ (coal), and even in some places and conditions to pure carbon as diamond. In Gold's view, there is good reason to consider hydrocarbon fuels as an almost unlimited source of energy

Gold gives strong arguments in favor of his views, using his outstanding erudition in astrophysics, chemistry, and biology. Some facts are impossible to explain by the biogenic theory:
-methane rivers now observed on the surface of cold planetary bodies like Titan, where a possibility of organic life never existed;
-tremendous methane outflow accompanying volcanic eruptions, together with hot lava; and
-refilling of depleted oil reservoirs, registered in many places, for instance near Eugene Island in the Gulf of Mexico (*Wall Street Journal*, 16 April 1999).

The main argument of biogenic theory proponents is the existence in crude oil of some molecules that indisputably belong to living matter because they manifest a chiral effect (predominant rotation of polarized light). The presence of biogenic molecules is actually an important part of the abiogenic concept as well, but it turns the old logic upside down. It is not that bacteria produced the hydrocarbons, but that the primordial hydrocarbon "soup" gave food to bacteria. The total mass of organic substance of these microbes is estimated to be hundreds of thousands of gigatons, much more than the organic mass of the surface biota.

Indeed, reasons exist to consider deep underground layers as the more probable place for the origin of life. Perhaps ten planetary bodies in our solar system could produce suitable subsurface abodes for the same kind of life as we have within Earth. Any rocky planetary body at least as big as our Moon might be expected to offer the requisite subsurface conditions of heat and upwelling hydrocarbons. An instrument placed on the Moon by an Apollo mission detected gas particles of atomic mass 16, which could only be methane. Panspermia (transfer of life) between planets of a planetary system would be a possibility, as the Martian meteorite has shown.

The triumph, not always recognized, of the abiogenic theory was the drilling experiment in the Siljan Ring in Sweden. Microbes were found at a depth of 6 km, eating oil and getting oxygen by reducing Fe_2O_3 to Fe_3O_4, thus leaving behind tiny particles of magnetite (fraction of a micron in size) as the product of metabolism. Gold himself observed that the liquid formed by these particles within the extracted oil sample had magnetic properties.

In addition to the drilling experiment in Sweden, a deeper one on the Kola peninsula in Russia had shown, in complete accord with Gold's predictions, that at a depth of about 10 km the fractured rocks were filled with highly compressed methane. Former life could not have reached such a depth and given such a tremendous quantity of methane.

Some tectonic movements might be connected with upwelling of methane and accompanying gases such as hydrogen sulfide, which are in turn connected with earthquakes. An increase of surface gas emissions before earthquakes is often sensitively detected by rats and other animals, who manifest unusual behaviour prior to earthquakes, as documented by numerous earthquake eyewitnesses.

Abiogenic theory also offers an explanation of the formation of metal deposits in the deep Earth. Leaching by upwelling of hydrocarbons creates along the way the organometallic compounds.

Gold respectfully acknowledges his predecessors in abiogenic theory, in particular the Russian and Ukrainian scientists Mendeleev, Koudriavtsev, Tchekaliuk, and Kropotkin. The last gave Gold much geological information.

Liquid methane cannot exist on Earth's surface due to its temperature. But, at high pressure methane might exist in the deep Earth on the upper mantle level in a supercritical state. One of the last arguments of biogenic advocates is: Yes, in Earth's crust it is possible to find abiogenic methane or other hydrocarbons, but the quantity is rather small, hence not commercial. What we see in the images of Titan is the large quantity, obviously not commercial for Earth 1.5 light hours away. The large quantities seen in volcanic eruptions, methane hydrates, or deep drilling as in the Kola peninsula are still not commercial either. Exploration for large quantities does not violate mass conservation or other physical laws. On Earth methane exists. In our view, the challenge is only for technology.

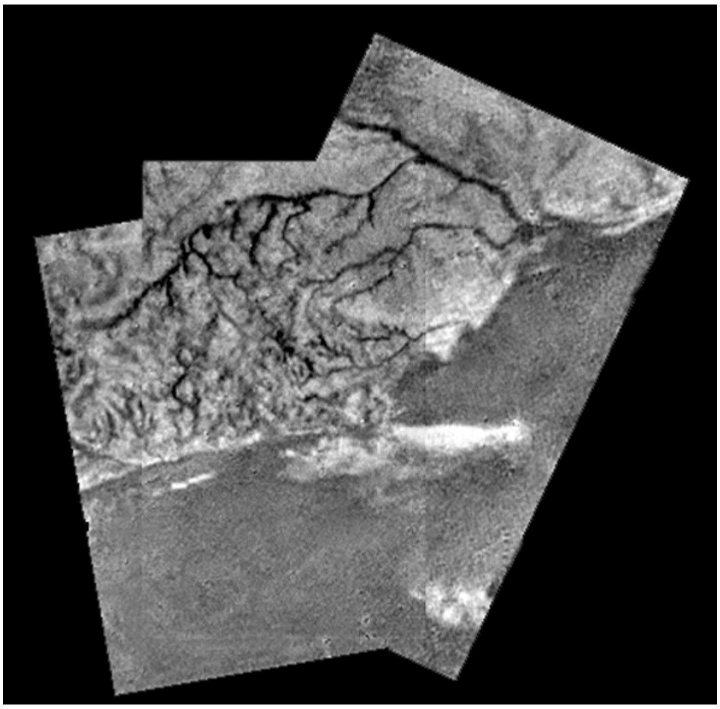

Figure 1. Rivers and sea of methane on the surface of Saturn’s moon Titan seen from an altitude of 8 km. Source: ESA.

Figure 2. Endless sea of methane photographed from Titan’s surface by probe Huygens, 14 January 2005 (ESA).

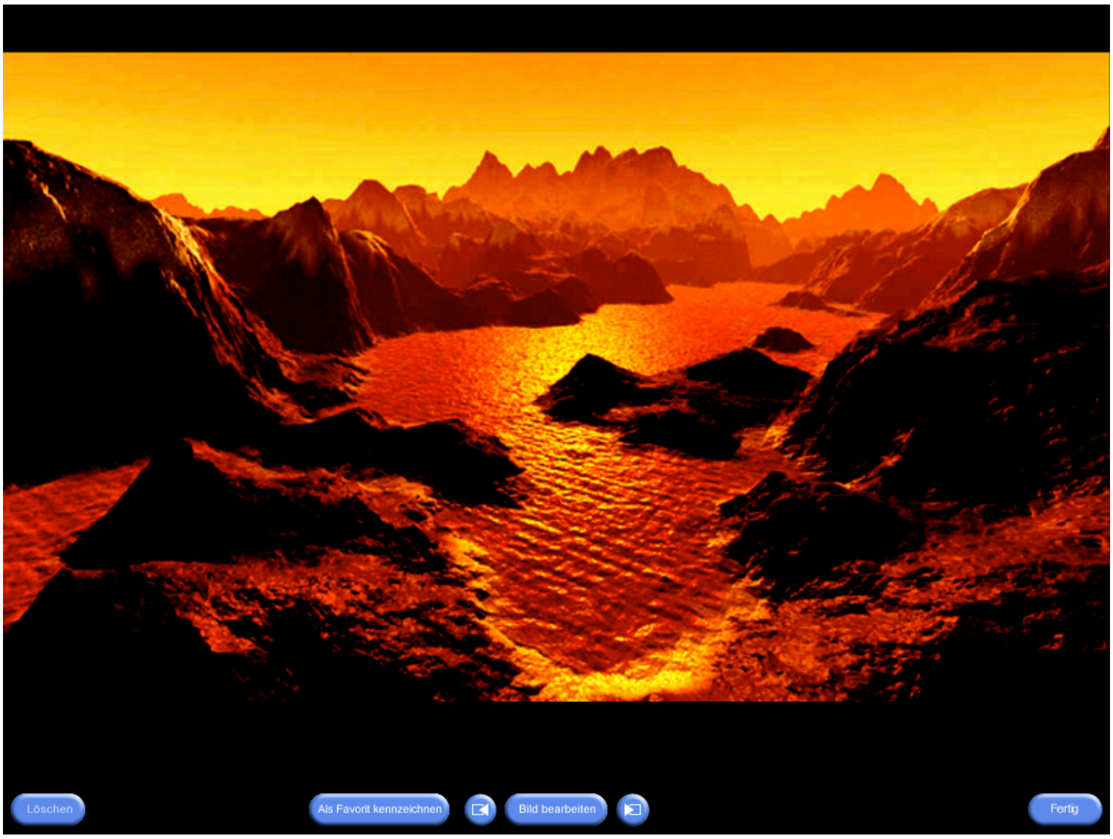

Figure 3. A lake of liquid methane surrounded by mountains of solid ice on Titan. Source: Huygens probe, ESA.

Evidence of methane production by inorganic processes has now also been shown experimentally by Scott et. al. who formed methane from FeO, $CaCO_3$-calcite, and water at pressures between 5 and 11 GPa and temperatures ranging from 500°C to 1,500°C (6). Other relevant data come from hydrothermal conditions (7). These experiments, backed by calculation, demonstrate that hydrocarbons may be formed in the upper mantle of Earth and confirm the general view formulated in a book by Korotaev et al. where P.N. Kropotkin compiled many data on abiogenic methane (8).

If sources of methane abound, the main restrictions on its use are pollution of the atmosphere by exhaust gases and the amount of oxygen available for combustion in the atmosphere. High-temperature, high-pressure zero-emission power plants (ZEPPs) offer the possibility of using currently available methane stores efficiently without pollution. As for the oxygen restriction, a shortage is unlikely to trouble the current century. The total mass of oxygen in the atmosphere is about 10^{15} tons, while the current consumption of oxygen is only about 3×10^{11} tons. Any problems caused by this 0.03% per annum reduction in oxygen levels have yet to become apparent. One hopes if they ever do become apparent, solar, nuclear, or other alternatives will have become the dominant sources of primary energy.

Thermodynamics of a reaction with methane formation of CO_2 and fayalite

Consider the simplified chemistry of carbon reactions (Equation 1) in a zero-emission power plant (above the line) and in an artificial geochemical reactor deep underground (below the line).

$$\begin{array}{ccccccccccc} CH_4 & + & & & 2O_2 & \Rightarrow & CO_2 & + & 2H_2O & + & \text{Power} \\ \hline \uparrow & & & & & & \downarrow & & & & \\ CH_4 & + & 4Fe_3O_4 & + & 6SiO_2 & \Leftarrow & CO_2 & + & 2H_2O & + & 6Fe_2SiO_4 \end{array} \tag{1}$$

The four arrows show a closed cycle of carbon. *Carbon plays the role of an energy carrier from fayalite to methane and then to the power plant.* At realistic conditions of temperature and pressure, some hundreds of degrees Celsius and some hundreds of bar, the reaction is exothermic and may be self-sustaining.

Below are reproduced some calculations by Akinfiev et al. (9) on formation of methane by a reaction of carbon dioxide with fayalite, an abundant mineral (10). *Fayalite becomes a fuel, with methane being the energy carrier. Viewed in this way, methane is a renewable energy source*, as theorized by Yantovski (11). Table 1 shows the properties of the reactants (12).

Table 1

Substance	Enthalpy ***h*, J/mol**	*Gibbs free energy* **g, *J/mol***
Fayalite	-1446764	-1424175
H_2O	-268156	-257755
$CO_{2,gas}$	-383990	-445257
Magnetite	-1078057	-1053163
Quartz	-898100	-868582
$CH_{4,gas}$	-65345	-95370

For the reaction

$$6Fe_2SiO_4 + 2H_2O + CO_{2,gas} = 4Fe_3O_4 + 6SiO_2 + CH_{4,gas} \quad (2)$$

properties of both liquid and/or gaseous H_2O were calculated using Hill's equation of state (13). At 250ºC and 200 bar we have

$$\Delta_r G_{250^o,\,200\,bar} = 4(-1053163) + 6(-868582) + (-95370) - 6(-1424175)$$
$$-2(-257755) - (-445257) = -13697 \text{ J} \quad (3)$$

$$\Delta_r H_{250^o,\,200\,bar} = 4(-1078057) + 6(-898100) + (-65345) - 6(-1446764)$$
$$- 2(-268156) - (-383990) = -165287 \text{ J} \quad (4)$$

The last value means that the reaction is exothermic.

The decimal logarithm of the thermodynamic constant of reaction for Equation 2 is

$$\lg K_a = \lg \frac{f_{CH4}}{f_{CO2}} = -\frac{\Delta_r G}{2.303RT} = -\frac{-14097}{2.303 \times 8.314 \times 523.15} = 1.37$$

Use of the Redlich-Kwong equation of state shows that the fugacity coefficients of the gases are close to 1 ($\gamma_{CO2} = 0.87$, $\gamma_{CH4} = 1.01$ at the T and P specified). This means that

$$\text{Log K} = 1.37 = \log P_{CH4}/P_{CO2} \quad (6)$$

$$\frac{p_{CH4}}{p_{CO2}} = 23.3 \quad (7)$$

The last value brings the conversion factor of CO_2 into CH_4 close to 96%.

We are unaware of experiments on the interaction of fayalite and carbon dioxide. However, the Albany Research Center extensively examined a similar reaction in an autoclave with comminuted (37 - 106 micrometers) forsterite Mg_2SiO_4 (14). Chemical energy of the forsterite is 256.8 kJ/mol. The reaction products are magnesite $MgCO_3$ and silica or silicic acid. The pressure was 117 - 127 atm and temperature 185 - 188°C. Test times spanned 3 to 48 h. A large increase of the reaction rate was observed when carbon dioxide was in a supercritical state. Similar experiments with comminuted fayalite might shed light on the kinetics and applicability of reaction (2) for production of methane.

Conclusion

Several diverse recent observations and experiments favor abiogenic theory and the possibility of finding deep in Earth what we see on the surface of Titan. Moreover, calculations show that the reaction between fayalite and CO_2 is exothermic and might be self-sustaining. A valuable next step would be experimental demonstration of the reaction of comminuted fayalite with carbon dioxide and water at conditions such as those mentioned above. Some better ‘fuels’ than fayalite might even be discovered. The search for methane in Earth and resolution of its origins deserve more efforts than ever before. In any case, we should pay tribute to Thomas Gold for his forecasts.

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