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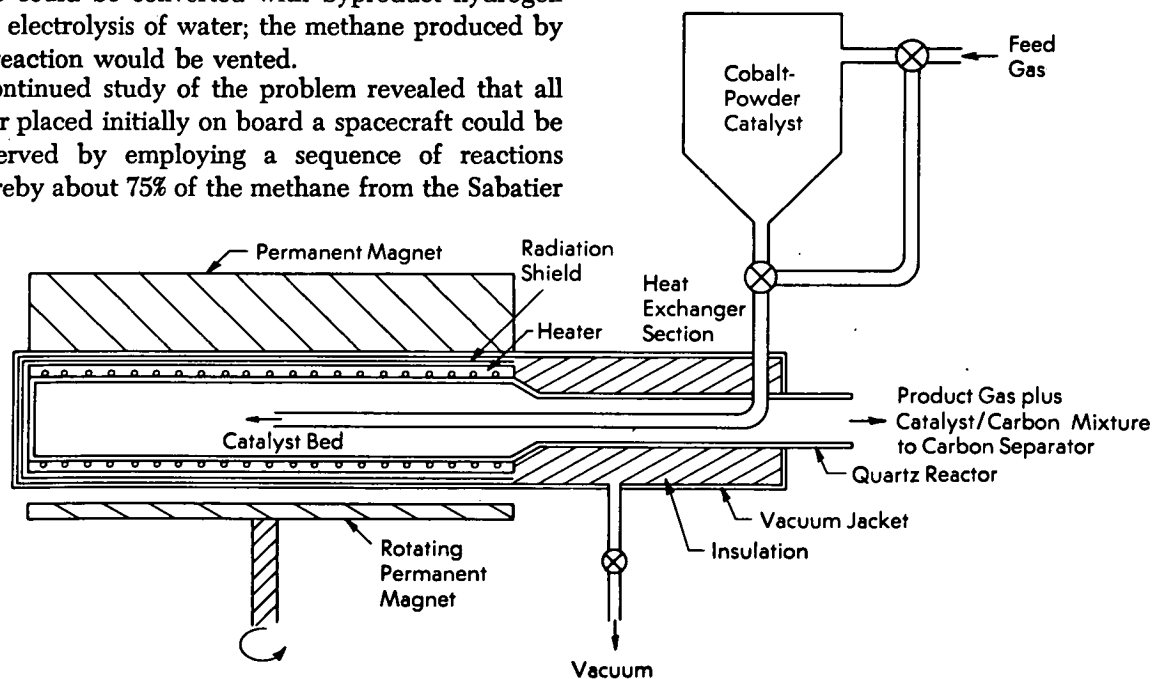
Continuous Catalytic Decomposition of Methane

The most reliable methods for producing oxygen in regenerative life support systems for space missions of long duration are based on the electrolysis of water. The need to minimize the amount of water which must be placed on board for a given mission led to a study of reactions which utilize hydrogen formed during electrolysis of water to convert metabolic carbon dioxide into water. Initial work indicated that the Sabatier reaction, which converts carbon dioxide and hydrogen into water and methane, appeared to be compatible with spacecraft requirements. However, the problem of water recovery was still unsolved because only about 60% of the metabolic carbon dioxide could be converted with byproduct hydrogen from electrolysis of water; the methane produced by the reaction would be vented.

Continued study of the problem revealed that all water placed initially on board a spacecraft could be conserved by employing a sequence of reactions whereby about 75% of the methane from the Sabatier

reaction is decomposed to solid carbon and hydrogen; the hydrogen is then separated from residual methane and utilized in the usual Sabatier reaction to reduce all the remaining metabolic carbon dioxide. In order to utilize the sequence of reactions, it was necessary to design a high-temperature (850°C) catalytic methane decomposition unit that weighs less than the onboard water it is to replace and can cope with the formation of solid carbon in a weightless environment.

The decomposition of methane in a nearly gravity-free environment can be accomplished with a powder-type metal catalyst if a magnetic field is used to suspend the catalyst in the gas stream within the hot



(continued overleaf)

reaction zone. The magnetic field within the hot reactor is established by external coils situated in a relatively cold environment and rapid fluctuations or reversals of the field strength can be used to agitate the catalyst particles and to provide a well-mixed bed. The catalyst particles are maintained in the reaction zone until sufficient utilization is achieved (carbon/catalyst ratio of 5 or higher); the resulting mixture of carbon and catalyst is removed from the reactor by entrainment in the gas stream (at reduced gravity) when the magnetic field is reduced in strength or removed. Removal of the carbon-catalyst mixture and feed of fresh catalyst can be continuous or periodic; the hydrogen formed in the reactor is recovered continuously.

The highest rates of reaction are obtained at the highest temperatures obtainable in the catalyst bed. However, to avoid the deposition of carbon on the walls of the reactor and to confine carbon to the catalyst surfaces, temperatures below 950°C are used; the preferred temperature is about 850°C. Only cobalt has a Curie point high enough to remain magnetic at 850°C. (The Curie point for cobalt is 1130°C, 770°C for iron and 358°C for nickel.) Alloy catalysts might also be useful (e.g., an alloy of cobalt with up to 40-percent nickel would have a Curie point above 850°C).

Large surface areas are needed to provide high reaction rates and, therefore, the catalyst must have a small particle size. However, a metallurgical structure that favors disintegration into even smaller particles by the wedging action of carbon formed in microcracks is very desirable. The cobalt catalyst used in the experiments was in the form of a powder (-50 mesh), and it was found that a preliminary activation of the catalyst provides greater reaction rates, e.g., cobalt powder oxidized in air at 800°C for one hour and reduced in hydrogen at 500°C for one hour.

Initial laboratory experiments have shown the feasibility of using a magnetic field to suspend, retain, and fluidize a cobalt powder catalyst bed in the hot reaction zone (850°C) for extended operating periods while methane is decomposed catalytically to carbon. Experiments have also provided preliminary design data for constructing the laboratory model illustrated in the diagram. The reactor for methane decomposition is one subsystem of the total system required to close the Sabatier reaction sequence. The other subsystem components are a filter assembly for final collection of spent carbon-catalyst mixture and a hydrogen-stripping unit. The hydrogen-stripping unit

consists of an electrochemical hydrogen concentration cell (Pd-25Ag hydrogen diffusion electrodes) that removes hydrogen anodically from the mixture of hydrogen and methane produced by the catalyst bed and cathodically generates an equivalent quantity of hydrogen for reaction with carbon dioxide according to the Sabatier reaction.

Notes:

1. The technology is not limited to the high temperature decomposition of methane, carbon-forming reactions, or catalytic reactions. For example, cobalt particles could be coated with a layer of functional material that might be used for adsorption of gases or vapor, and possibly in a regenerative system where the bed is moved magnetically from a cold to a hot adsorption zone. Catalytic reactions in beds supported by magnetic fields are attractive because the detrimental channeling of gas that inevitably occurs in packed beds is avoided. Moreover, magnetically supported beds are excellent alternatives when fluidized-bed techniques cannot be used. The magnetically supported catalyst beds may be used in any gas-phase catalytic reaction (such as in the Bosch system) or for disproportionation reactions in solid electrolyte systems which use high-temperature reactors to produce carbon.
2. The following documentation may be obtained from:

National Technical Information Service
Springfield, Virginia 22151
Single document price \$6.00
(or microfiche \$0.95)

Reference:

NASA CR-1662 (N70-42522), Continuous Catalytic Decomposition of Methane.

3. No additional documentation is available. Specific questions, however, may be directed to:

Technology Utilization Officer
Ames Research Center
Moffett Field, California 94035
Reference: B73-10016

Patent status:

NASA has decided not to apply for a patent.

Source: John E. Clifford, Byung C. Kim, Janez Zupan, Louis J. Hillenbrand, and Edwin S. Kolic of Battelle Memorial Institute—Columbus Laboratories under contract to Ames Research Center (ARC-10339)