Preliminary Experimental Results of Gas Recycling Subsystems Except Carbon Dioxide Concentration


* Mitsubishi Heavy Industries, Ltd. 10, Oye-cho, Minato-ku, Nagoya, 455, Japan. ** Mitsubishi Heavy Industries, Ltd. 2-1-1, Shinshama, Arai-cho, Takasago, 676, Japan. *** Kawasaki Heavy Industries, Ltd. 3-1-1, Higashikawasaki-cho, Chu-ku, Kobe, 650-91, Japan. ++ National Aerospace Laboratory, 1880, shindaiji-cho, Chofu, Tokyo, 182, Japan.

Abstract

Oxygen concentration and separation is an essential factor for air recycling in a CELSS. Furthermore, if the value of the plant assimilatory quotient is not coincident with that of the animal respiratory quotient, the recovery of O₂ from the concentrated CO₂ through chemical methods will become necessary to balance the gas contents in a CELSS. Therefore, oxygen concentration and separation equipment using Salcomine and O₂ recovery equipment, such as Sabatier and Bosch reactors, were experimentally developed and tested.

1. Basic Consideration on Gas Recycling

Fundamental functions of gas recycling in CELSS, as shown in Fig. 1.1, are to separate each component, such as O₂, CO₂, and N₂ gases, and to concentrate and store each gas in order to supply appropriate gas concentration for human beings, plants, and algae.

2. Oxygen Recovery from Concentrated Carbon Dioxide

Many research and development efforts have been conducted for the recovery of oxygen from concentrated CO₂ by the catalytic hydrogenization process. The Bosch reaction utilizes an iron catalyst and produces carbon (C) and water (H₂O) with CO as an intermediate product /1/. Product water is further electrolyzed to recover O₂ for animal respiration and H₂ for subsequent hydrogenization. The Sabatier reaction utilizes a ruthenium (Ru) catalyst and produces methane and water /2/. An additional CH₄ cracking process providing C and H₂ is necessary to make the Sabatier reaction comparable to the Bosch reaction /3/.

Mitsubishi Heavy Industries, Ltd. (MHI) has been involved in CELSS research for several years /4/ under the direction of the National Aerospace Laboratory and conducted the experimental program to evaluate the basic characteristics and performance of the two oxygen recovery processes.

2.1 Once Through Tests

In order to determine the basic characteristics of each reaction, once through tests of CO₂ reaction with H₂ on the catalyst are performed. Fig. 2.1 shows the schematic of the test set up.

Bosch Reaction Fig. 2.2 shows the reaction rate of CO₂ vs. reaction temperature in the first Bosch reaction, as well as the reaction rate of CO vs. temperature in the second Bosch reaction. The different nature of the two reaction rate curves suggests a phased process concept using two reactors operating at different temperature ranges to obtain higher performance in the Bosch CO₂ cracking process.

---

Fig. 1.1 Fundamental function of Gas Recycling System

Fig. 2.1 Bosch/Sabatier Once Through Test Setup
2.2 Reaction Efficiency vs Temperature of Bosch Reaction

Sabatier Reaction: Fig. 2.3 shows the reaction rate of $\text{CO}_2$ vs. mole ratio of $\text{H}_2/\text{CO}_2$ in the feed gas mixture of the first Sabatier reaction. Reaction rate of more than 99% is achieved with a little bit higher ratio (4.5) than the stoichiometric value. As for the second Sabatier reaction, reaction rate of $\text{CH}_4$ vs. time after start of the reaction is presented in Fig. 2.4. Very rapid degradation is observed for the catalytic reactions using Pt or Ni, while a steady and much higher conversion efficiency is observed in pyrolytic reaction on silica wool filler.

2.2 Recycle Test

A Recycle Test Apparatus was prepared based on the once through test results as shown in Fig. 2.5.

The supply rate of mixture gas is shown in Fig. 2.6. The amounts of processed $\text{CO}_2$ are 0.15 kg/day and 0.42 kg/day for Bosch and Sabatier, respectively. Energy required for the above processes were estimated and the Sabatier process showed several times less energy use than Bosch process. The character of deposit carbon in the Bosch process was a loose powder form, while a hard solid block was obtained from the Sabatier reactor. This fact has an important meaning for maintenance operations to periodically extract carbon on orbit.

Thus, the $\text{O}_2$ recovery system with Sabatier methane cracking shows a good possibility for application in CELSS and Space Station.
3. OXYGEN SEPARATION SYSTEM USING SALCOMINE

3.1 Functions of Oxygen Separation System

Kawasaki Heavy Industries Ltd. (KHI) has been involved in CELSS research under contract with the National Aerospace Laboratory (NAL) /5/.

Fig. 3.1 shows the function of O$_2$ separation and concentration system, concerning the CO$_2$ and N$_2$ separation and concentration. In the Fig. 3.1, the inlet gas is mixture of O$_2$, CO$_2$ and N$_2$. At the CO$_2$ concentrator, CO$_2$ is separated and concentrated, and stored in the CO$_2$ gas bottle. At the O$_2$ concentrator, O$_2$ is separated and concentrated, and stored in the O$_2$ gas bottle. The residual is N$_2$ gas and this is stored in the N$_2$ gas bottle. Thus, inlet gas is separated into three (3) components, CO$_2$, O$_2$, and N$_2$ having high purities.

There are many methods for CO$_2$ separation and concentration. The studies reported here are focused on the O$_2$ separation and concentration system using Salcomine. Salcomine absorbs the O$_2$ under normal temperature (lower than about 40°C) and desorbs the O$_2$ under high temperature (higher than about 80°C). So, when the inlet gas is introduced into the Salcomine canister, the included O$_2$ is absorbed by Salcomine. After the Salcomine absorbs the O$_2$, the Salcomine canister is heated and Salcomine desorbs O$_2$ of high purity.

In the Fig. 3.1, there are three (3) canisters and each canister is operated in absorbing, desorbing and pre-cooling modes respectively. Thus, continuous O$_2$ separating operation can be carried out by the cyclic exchange of these operation modes.

3.2 SALCOMINE

Fig. 3.2 shows the structural formula of the Salcomine and its O$_2$ absorption and desorption. There are some studies on the application of Salcomine for O$_2$ concentration. Natsuda /6/ carried out the experimental study on the O$_2$ absorbing and desorbing characteristics and their variations under repeated reaction of the Salcomine, as a part of the development of Diffusive Atmosphere Control System (DACS, a kind of artificial gill), which extract O$_2$ from sea water dissolving oxygen.

The present study is based on these results and is designed to get data to determine the optimum O$_2$ separating and concentrating system for CELSS. In this study, O$_2$ absorbing and desorbing performance tests of Salcomine within the canister are carried out to get the data to design a compact, light weight and lower energy consuming system.

3.3 Oxygen Absorbing and Desorbing Test of SALCOMINE

3.3.1 Test of SALCOMINE Proper.

In Fig. 3.3(a), oxygen containing nitrogen gas is introduced into the equipment and Salcomine absorbs the O$_2$, and increases its weight. And then the Salcomine is heated and the Salcomine decrease its weight desorbing O$_2$. From the weight change of the Salcomine, the O$_2$ absorbed and desorbed is measured.

Test results are shown in Fig. 3.3(b). O$_2$ absorbing and desorbing capacity of Salcomine is expressed with the percentage of oxygen weight to the Salcomine weight (wt%). From Fig. 3.3(b), O$_2$ absorbing capacity is about 4.0 wt% for 20% of O$_2$ concentration.

3.3.2 Test of SALCOMINE within Canister.

In Fig. 3.4, O$_2$ containing air is drawn by the air pump and introduced into the Salcomine canister. In the canister, O$_2$ in the air is absorbed by Salcomine.

Fig. 3.2 Oxygen Absorb and Desorb Reaction by SALCOMINE

(a) Test Equipment (b) Test Results

Fig. 3.3 Characteristic Test of SALCOMINE Proper
And at the exhaust gas manifold, O₂ concentration is measured. During this time, the Salcomine canister is cooled. For the desorbing test, the canister is heated by hot water (90°C) and drawn by the vacuum pump. O₂ desorbed from the Salcomine is collected into the sampling bag and analysed.

Fig. 3.5 shows the results of the test. Initially, O₂ concentration in the outlet air is almost 0% because all O₂ in the air is absorbed. After a few minutes, the break-through occurs and O₂ concentration rises, and reaches to 21% which is equal to that of the inlet air. From Fig. 3.6, absorbed O₂ is 675CC and this corresponds to 2.7 wt% of O₂ absorbing capacity. On the start of O₂ desorbing test, the vacuum pump is operated to purge the air inside the test equipment. The canister is then heated. And is drawn by the vacuum pump. The drawn gas is measured 710 CC, and O₂ concentration in this gas is measured 51.4%. So, desorbed O₂ becomes 649 CC.

4. CONCLUSION

As a part of CELSS Gas Recycling System Development, preliminary experimental studies were conducted for the processes of O₂ recovery from concentrated CO₂ and of O₂ separation and concentration from exhaled gas of animal, plant etc. Sabatier process for the regenerable O₂ recovery from CO₂ shows a good possibility compared with Bosch process in its higher efficiency of O₂ recovery and easier handling of product carbon. From the O₂ separation and concentration tests using Salcomine, oxygen absorbing and desorbing characteristic data are obtained as the fundamental design data. The above results can be used in the planning of a complete and stable gas recycling system required in CELSS. We hope to continue our research and development effort for the expansion of the human frontier in Space.

REFERENCES