STUDY ON A HYDROGEN-REDUCED REACTOR DESIGN FOR LUNAR WATER PRODUCTION

Sachio Ogiwara¹, Kunihisa Eguchi², Hiroshi Kanamori³, Hideto Yoshida⁴, and Takayuki Watanabe⁵

Abstract

Our research group has been conducting a basic work on experimental missions for lunar resource utilization since 1996. The objective of the research work is to provide a conceptual design of the In-situ Space Resource Utilization (ISRU) experiment system for unmanned water production on the Moon, and to define essential technological breakthroughs. As part of the research program, an experimental study on the hydrogen reduction of lunar soil has been performed to design a chemical reactor for water production.

Keywords: moon, resource utilization, water production, chemical reactor, hydrogen reduction, lunar-soil simulant.

Introduction

Progress in the technologies for producing and utilizing lunar-derived materials will be vital by the time of manned lunar-stay missions [1]. In particular, water is considered to be important among these materials, because it is required for life support, regenerative fuel cells with electrolyzers, thermal management, and various chemical processes. In addition, the water-production process involves essential technologies for resource utilization, such as the reduction and direct use of solar-heat energy. Therefore, water-production technology will be definitely required on the moon, particularly until water is surely provided from the lunar polar region or other source.

In the process of water production, the iron oxide contained in lunar soil is reduced with hydrogen. This reaction is endothermic with 11 kJ/mol under 1300 K, and can be

¹ Senior Researcher, Space Project and Research Center, National Aerospace Laboratory.
7-44-1, Jindaiji Higashi, Chofu, Tokyo 182-8522, Japan
ogiwara@nal.go.jp
Tel +81-422-40-3152 Fax +81-422-40-3146
² Leader, Stratospheric Platforms Research Group, Fluid Science Research Center, National Aerospace Laboratory.
7-44-1, Jindaiji Higashi, Chofu, Tokyo 182-8522, Japan.
eguchi@nal.go.jp
Tel +81-422-40-3492 Fax +81-422-40-3441
³ Manager, Space Systems, Shimizu Corporation.
1-2-3, Shibaura, Minato, Tokyo 105-8007, Japan.
kanamori@sps.shimizu.co.jp
Tel +81-3-3820-5455 Fax +81-3-3820-5392
⁴ Graduate School Student, Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology.
2-12-1, Ookayama, Meguro, Tokyo 152-8550, Japan.
99m21336@nr.titech.ac.jp
Tel +81-3-5734-3058 Fax +81-3-5734-2959
⁵ Associate Professor, Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology.
2-12-1, Ookayama, Meguro, Tokyo 152-8550, Japan.
watanabe@nr.titech.ac.jp
Tel +81-3-5734-3058 Fax +81-3-5734-2959
expressed by a chemical reaction:

$$\text{FeO(s)} + \text{H}_2(\text{g}) \Rightarrow \text{Fe(s)} + \text{H}_2\text{O(g)}$$  (1)

Since the change in Gibbs’ free energy in this reaction is relatively small, the reduction of FeO can easily occur, which is necessary for lunar water production.

Based on the ideas mentioned above, a water-production experiment mission was conceptually designed by our research group, and fundamental specifications of subcomponents, such as reactor, solar furnace, measurements and analyzer, and sample handling device, were first defined [2]. The present objective was to define the detailed specifications of the reactor, a key component of the overall system. For this purpose, significant information on the reactor design was obtained from our experiments using a fixed-bed reaction test model and lunar soil simulants. This paper will describe the outline of the experimental study.

Experiments

Parameters

Parametric conditions for our experiments are shown in Table 1. The experiments were programmed to examine three different parameters, which were sample quantity, reaction temperature, and sample particle size.

**Table 1 Test conditions.**

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quantity of lunar soil simulant</td>
<td>40 g</td>
</tr>
<tr>
<td></td>
<td>70 g</td>
</tr>
<tr>
<td></td>
<td>100 g</td>
</tr>
<tr>
<td>Reaction temperature</td>
<td>1173 K (900°C)</td>
</tr>
<tr>
<td></td>
<td>1223 K (950°C)</td>
</tr>
<tr>
<td></td>
<td>1273 K (1000°C)</td>
</tr>
<tr>
<td></td>
<td>1298 K (1025°C)</td>
</tr>
<tr>
<td></td>
<td>1323 K (1050°C)</td>
</tr>
<tr>
<td>Particle size (Median size)</td>
<td>150 μm</td>
</tr>
<tr>
<td></td>
<td>120 μm</td>
</tr>
<tr>
<td></td>
<td>90 μm</td>
</tr>
<tr>
<td></td>
<td>60 μm</td>
</tr>
<tr>
<td></td>
<td>30 μm</td>
</tr>
<tr>
<td></td>
<td>70 μm (Sall)</td>
</tr>
</tbody>
</table>

**Fixed-bed Test Apparatus**

A schematic of the test apparatus is illustrated in Figure 1. The apparatus consists of a reactor, a furnace, and a measurement line including a moisture meter, gas flow meters, pressure gauges, thermocouples, an A/D converter, and a personal computer for data acquisition. In the figure, the letters ‘T’ and ‘V’ indicate thermocouples and valves, respectively.

Figure 2 shows a cross-sectional view of the reactor. It is made of Inconel-600, and has an annular passage, which consists of an inner tube of 30 mm i.d. and 275 mm long, and an outer tube of 46 mm i.d. and 265 mm long. The soil sample is held in the upper
part of the inner tube by placing ceramic screen filters with 10 \( \text{nm} \) openings and glass wool on the top and bottom ends of the sample. Hydrogen gas comes from an inlet of the outer tube, flows up through a hydrogen-preheating gap between the inner and the outer tubes, and goes into the sample through the upper filter. A mixture of reacted and unreacted gases is sent to the moisture probe after the outlet. A split-cylinder-type kanthal-wired heater was used for the furnace, and ribbon heaters were wrapped around the pipes downstream of the reactor to prevent water condensation. A capacitance-change-type probe was used for moisture measurements.

\* T10 and T11 control the temperature of ribbon heaters. T12 controls temperature of the saturator.

**Figure 1** Schematic of fixed-bed test apparatus.

**Figure 2** Cross-sectional view of reactor tubes.
Solid Samples

As shown in Table 2, six types of lunar soil simulant are selected as solid samples for the reduction tests. An original type of sample, named FJS-1, is manufactured from basaltic lava rocks in Japan. In our testing, the original sample is referred to as “Sall”, and is screened in different ranges of particle size to produce five other sample types.

<table>
<thead>
<tr>
<th>Types</th>
<th>Size range (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S150</td>
<td>135 - 165</td>
</tr>
<tr>
<td>S120</td>
<td>105 - 135</td>
</tr>
<tr>
<td>S90</td>
<td>75 - 105</td>
</tr>
<tr>
<td>S60</td>
<td>45 - 75</td>
</tr>
<tr>
<td>S30</td>
<td>15 - 45</td>
</tr>
<tr>
<td>Sall</td>
<td>1 - 2000</td>
</tr>
</tbody>
</table>

The mechanical properties of the Sall type are specified below.

Specific gravity: 2.94
Particle size: 1 – 2000 μm (median size: 70 μm)
Minimum bulk density: 1.55 g/cm³
Cohesion c: 8 kPa
Internal friction angle φ: 37.2 deg.

Test Procedure

The present tests were run using the following procedure.

(i) Chemical composition analysis of the solid samples with X-ray fluorescence.
(ii) Sample weighing and packing into the reactor.
(iii) Sealing of the reactor.
(iv) Preparation for the reduction test.

Heaters on.
Letting helium flow through the reactor.
Bypassing hydrogen at a flow rate of about 2 liter/min.
Starting data acquisition.

(v) Starting reaction.
Change of helium to hydrogen flow.
Measure moisture, temperatures, flow rate, and pressures.

(vi) Sample recovery, weighing, and analysis.

Results and Discussions

Chemical Composition of Solid Samples

The measured chemical compositions of the samples are shown in Table 3. All the samples used in our test are considered to have almost the same chemical constituents, regardless of the different particle size of each sample. As far as the effect of reduction, a
reacted sample of the Sall type showed an increase in FeO content and a decrease in Fe₂O₃ content. Since a method concerned for the quantitative measurement of FeO in our test can not determine the FeO and Fe contents separately, FeO is expected to contain a large amount of Fe.

Table 3 Chemical composition of lunar soil and test samples.

<table>
<thead>
<tr>
<th>Components</th>
<th>Apollo lunar samples (mean)</th>
<th>Lunar soil simulators</th>
<th>Sall* before</th>
<th>after</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>11</td>
<td>12</td>
<td>14</td>
<td>15</td>
</tr>
<tr>
<td>SiO₂</td>
<td>42.2</td>
<td>46.3</td>
<td>48.1</td>
<td>46.9</td>
</tr>
<tr>
<td>TiO₂</td>
<td>7.8</td>
<td>3.0</td>
<td>1.7</td>
<td>1.4</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>13.6</td>
<td>12.9</td>
<td>17.4</td>
<td>14.6</td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>0.3</td>
<td>0.3</td>
<td>0.2</td>
<td>0.4</td>
</tr>
<tr>
<td>FeO</td>
<td>15.3</td>
<td>15.1</td>
<td>10.4</td>
<td>14.3</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>MnO</td>
<td>0.2</td>
<td>0.2</td>
<td>0.1</td>
<td>0.2</td>
</tr>
<tr>
<td>MgO</td>
<td>7.8</td>
<td>9.3</td>
<td>9.4</td>
<td>11.5</td>
</tr>
<tr>
<td>CaO</td>
<td>11.9</td>
<td>10.7</td>
<td>10.7</td>
<td>10.8</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.5</td>
<td>0.5</td>
<td>0.7</td>
<td>0.4</td>
</tr>
<tr>
<td>K₂O</td>
<td>0.2</td>
<td>0.3</td>
<td>0.6</td>
<td>0.2</td>
</tr>
<tr>
<td>P₂O₅</td>
<td>0.1</td>
<td>0.4</td>
<td>0.5</td>
<td>0.2</td>
</tr>
<tr>
<td>S</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.1</td>
</tr>
<tr>
<td>H₂O</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Total</td>
<td>100.0</td>
<td>99.0</td>
<td>99.8</td>
<td>101.0</td>
</tr>
</tbody>
</table>

* Note: Before and after the reaction

Effect of Sample Quantity

Figure 3 shows the relationship between the sample quantity and the total amount of water produced during the reduction process. A linear line as drawn in Figure 3 indicates a proportional relationship between the yield water and sample quantity. Since the value for 40g-sample is plotted slightly below the line, it is estimated that the mean temperature of the 40g-sample during the reaction is a little lower than that of the other samples.

Thus, the reactor will be subjected to a temperature gradient in the vertical direction.

![Figure 3 Sample quantity and yield-water amount.](image)

Effect of Reaction Temperature
The total amounts of yield water for different reaction temperatures are plotted in Figure 4. For the temperatures below 1273 K, the amount of the produced water increased with elevating of the temperature. To gain high reactivity for much more water production, the reaction temperature should be generally kept as high as possible. When the reaction temperature exceeds 1273 K, however, the amount of water produced tends to decrease with increasing temperature. The cause is considered to be partial sintering of the sample. Since the hydrogen gas does not reach the sintered area, some portion of FeO or Fe₂O₃ is left unreacted in the sample. Consequently, it is found from our experiment that the temperature of 1273 K provides an optimum reaction process to obtain higher reactive efficiency.

**Figure 4 Total yield-water vs. reaction temperature.**

**Effect of Sample Particle-Size**

**Figure 5 Changes of yield-water mass for different particle sizes.**

Figure 5 shows time-dependent variations of the produced water mass for six test samples of different particle-size. In general, the reaction time to reach the peak rate of water production is approximately 100 seconds, and the peak value differs depending on the particle size. The
sample of S30 type has a peculiar tendency, requiring twice as much time to complete the reaction than the other samples. The major reason for it is that the hydrogen flow rate is relatively low for this case. The hydrogen flow decreases because fine particles like the S30 type form a high-density-packed bed in the reactor. As a result, it is observed that the hydrogen mass transfer process is one of the key water-production processes to control reaction rates of the selected samples.

Significant Considerations to Water Production Reactor Design

Reactor

(i) The size and material of the cylindrical reactor vessel used in this study will be applicable to an actual reactor design for a lunar water-production mission. The reactor has a capability of reducing lunar soil of approximately 100 g with hydrogen.

(ii) A preferable reaction temperature is 1273 K, assuming that the sintering temperature of the actual lunar soil is quite the same as that of the lunar soil simulant used in this study.

(iii) The amount of hydrogen to reduce lunar soil simulant of 1 g needs about $3 \times 10^4$ m$^3$.

Furnace

(i) The furnace contributes to homogeneous heating of lunar sample in the reactor.

(ii) Thermal control of the furnace is required to avoid over-heating or sintering of the lunar sample.

Conclusions

In conclusion, the most significant findings are summarized as follows.

(i) All simulants used in our tests have almost the same chemical compositions regardless of the difference in particle size.

(ii) FeO and Fe$_2$O$_3$ contained in the simulant are major substances to be reduced by hydrogen.

(iii) A hydrogen reduction at a reaction temperature of 1273 K produces relatively high amounts of water compared with the reductions at other temperatures.

(iv) A mass-transfer behavior of the hydrogen flow around solid particles is one of the key hydrogen-reduction factors to control the reaction rate.

(v) The internal portion of the relatively large solid particle is partially left unreacted.

Because of the limited test conditions, such as hydrogen flow rate and pressure, further modifications of the experimental apparatus will be needed to examine more details of the reaction process, and to meet completely the reactor design requirements.

References
