

## 8 / OXYGEN: PRELUDE TO LUNAR INDUSTRIALIZATION

**P**ROJECT APOLLO WAS INITIATED and then truncated by the political process in the United States. Very probably the next manned landings on the Moon will occur as the result of political decisions. As long as political motivations are the principal drivers for lunar activity, then realignments of national priorities also can terminate future programs.

In 1969, the Apollo flights were the only manned space activity. In the 21st Century, lunar surface activities will be an extension of a manned operations in near-Earth space, including routine flights between Earth and LEO and continuous presence in one or more LEO space stations. Although future flights to the Moon will not seem as exotic as they once did, lunar operations will not be secure until the surface of the Moon is viewed not as an outpost but as an integral part of the space infrastructure. To achieve this end, the lunar activity must return some benefit to the system.

For the foreseeable future, large scale operations in space will be hampered by the large energy required to launch from the surface of the Earth. Payloads can be launched from the Moon to LEO with an energy expenditure that is more than an order of magnitude less. Although hypothetical lunar products might have a lower transportation cost, the capital investment required for establishing and operating lunar manufacturing facilities might well make the concept infeasible. Two lunar products with potentially large markets yet requiring minimal processing are simple regolith for use as shielding mass and liquid oxygen for spacecraft propellant.

Production of oxygen propellant is attractive because it immediately relieves some of the burden on the transportation system for lunar operations. To place one kilogram on the lunar surface requires launching approximately seven kilograms to LEO, most of which is fuel for the trip. A small production plant capable of fueling a reusable vehicle shuttling between the base and lunar orbit halves the required launch mass to LEO. If the plant produces enough oxygen for the whole

transportation system, then only approximately 1.2 kilograms are required from the Earth to send one kilogram on to the Moon.

A lunar production facility capable of supporting lunar operations and supplying excess propellant to LEO appears feasible. Simon addresses the key question whether the price of lunar oxygen in LEO can be less than that for propellant shipped from Earth. Although an accurate answer cannot be given at the present time, Simon's methodology explicitly identifies the relevant parameters and points to key technologies where research is needed.

An important element of the total system analysis is the process chosen to produce the oxygen. Several are possible, but the reduction of the mineral ilmenite by hydrogen has been studied somewhat more than others. Gibson and Knudsen discuss the process and potential reactor designs. Williams presents experimental data on ilmenite reduction. Cutler and Krag describe an alternative process that could be easily augmented to produce steel for use on the Moon or as an export for space construction.

A major irreducible cost element in production of lunar oxygen is the shipping cost of imported hydrogen. Whether hydrogen of solar wind origin can be recovered from the lunar regolith becomes an interesting technical issue. Carter constructs a simple model of the hydrogen rich layer on lunar grains. Tucker *et al.*, explore the use of microwave energy to free hydrogen in the soil thermally. White and Hirsch suggest the possibility of microbial processing of lunar regolith to extract absorbed volatile gases. Blanford *et al.*, have implanted ions into minerals to determine experimentally the behavior of gases embedded in the crystal lattice.

Friedlander suggests the delivery of hydrogen to the Moon in the form of methane or ammonia. Storage and handling of these compounds is much simpler than for liquid hydrogen, and the critical elements carbon and nitrogen can be made available for life support systems.

# A PARAMETRIC ANALYSIS OF LUNAR OXYGEN PRODUCTION

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A methodology has been developed for evaluating alternative approaches to utilization of space resources. The specific application discussed in this paper is a parametric cost analysis of liquid oxygen (LO<sub>2</sub>) production on the Moon. A baseline scenario for production of LO<sub>2</sub> from lunar ore and its delivery to low Earth orbit (LEO) is defined, and the major systems required to support such an enterprise are identified. Fifteen principal variables influencing the capital emplacement and operations costs are defined and organized into a parametric cost model. A sensitivity analysis is performed to identify the impact of each of these variables on the costs of lunar LO<sub>2</sub> production. The cost model and sensitivity analysis are structured with emphasis on permitting evaluation of alternatives to the baseline scenario, rather than for developing conclusive cost estimates at the present time. Economic weighting factors are derived to provide a measure of the potential impact on cost from each of a number of key technologies. Using these factors, certain types of near-term technology development activities are recommended to maximize future cost-effective utilization of space resources.

## INTRODUCTION

Utilization of space resources, *i.e.*, raw materials obtained from non-terrestrial sources, has often been cited as a prerequisite for large scale industrialization and habitation of space. While transportation of extremely large quantities of material from Earth would be costly and potentially destructive to our environment, vast quantities of usable resources might be derived from the Moon, the asteroids, and other celestial objects in a cost-effective and environmentally benign manner.

Of more immediate interest to space program planners is the economic feasibility of using space resources to support near-term space activities, such as scientific and commercial missions in the 2000–2010 time frame. The purpose of the analysis described in this paper is to identify the factors most likely to influence the economics of near-term space resource utilization, based on development of a baseline scenario for liquid oxygen production from lunar ore. Liquid oxygen as a propellant for space-based transportation systems appears to be an obtainable space resource with the firmest near-term requirement for quantities sufficient to be produced economically in an extraterrestrial setting.

## ANALYSIS METHODOLOGY

The primary purpose of the parametric cost model developed as part of this study is to identify the factors that have the greatest influence on the economics of space

resource utilization. In the near term, this information can be applied to the establishment of technology development strategies, so that space resource utilization capabilities are developed in a manner likely to achieve cost-effective results.

It is important to note that predicting the actual costs of particular space resource utilization scenarios is only a secondary objective of this analysis. Estimates are made and dollar values are assigned principally as a relative measure of alternative options. Since the technologies for space resource utilization are in an early stage of development, it is premature to state conclusively whether mining the Moon, asteroids, or other celestial bodies makes economic sense. For these reasons the parametric model is designed more for flexibility than for precision. Although preliminary estimates indicate that production of oxygen from lunar ore is one of the space resource projects most likely to yield an economic payback, this activity was selected as a "baseline scenario" primarily because the requirements for such an endeavor can be relatively well defined. Hence, identification of the major systems required to support the baseline scenario could be made without much difficulty as being (1) a processing and storage facility to manufacture liquid oxygen from lunar ore and store it on the Moon, (2) a system of habitable lunar base elements to support a small, full-time crew, (3) a power system to provide the energy required for processing and storing the LO<sub>2</sub> and to support other lunar operations, and (4) a transportation and logistics system to deliver and support lunar base elements and to transport the lunar LO<sub>2</sub> to LEO.

Once these major support systems were defined, 15 key variables were identified as influencing the costs of developing and operating these systems (Table 1). Cost variables were limited to general categories so that the parametric model could be adapted in the future to evaluation of alternative scenarios. Next, equations were developed to calculate capital and operations costs as functions of these variables. Utilizing the codes and units detailed in Table 1, these equations are as follows:

$$\text{Capital cost} = (p \times c_p) + (n_t \times c_n) + (n_m \times c_u) + c_f + c_t \times [(p \times m_p) + (n_m \times m_m) + m_f]$$

$$\text{Operations cost} = c_t \times [(n_r \times m_m) + (1 - d) \times (125,000)] + (n_b \times n_f \times \$100,000)$$

where the capital cost is defined as the total cost of developing, building, and installing the lunar base systems (including transportation costs), and the operations costs are the annual costs of manufacturing 1 million kg (1,000 metric tons) of LO<sub>2</sub> per year and delivering to LEO as much of this LO<sub>2</sub> as possible.

The first two constants that appear in the operations cost equation reflect the assumptions that a portion of the LO<sub>2</sub> produced on the Moon is utilized as propellant to deliver the remaining LO<sub>2</sub> to LEO, and that 1 kg of hydrogen must be delivered from Earth to the Moon for every 8 kg used as propellant en route from the Moon to LEO. A higher-than-usual mixture ratio of 8:1 was selected for the baseline case after initial analyses showed the resultant reduction in hydrogen requirements to offer substantial economic benefits. The third constant in the operations cost equation is the cost per

Table 1. Lunar Oxygen Production—Major Cost Variables

Variable	Code	Units of Evaluation
Power required	p	Megawatts of installed capacity
Cost of power	$c_p$	Non-recurring cost per megawatt of installed capacity
Number of types of lunar base modules	$n_t$	Number of units
Cost of modifying space station modules	$c_n$	Non-recurring cost for adapting each type of module (\$)
Number of lunar base modules	$n_m$	Number of units
Unit cost of lunar base modules	$c_u$	Recurring cost of producing each lunar base module (\$)
Processing/storage facility cost	$c_f$	Development and production cost (\$)
Power system mass	$m_p$	Kilograms per megawatt of installed capacity
Earth-to-Moon transportation cost	$c_t$	Cost (\$) per kilogram delivered from Earth to the Moon
Mass of lunar base modules	$m_m$	Kilograms
Mass of processing/storage facility	$m_f$	Kilograms
Number of lunar base resupply missions/year	$n_r$	Number
Net lunar oxygen delivered to LEO	d	Fraction of lunar $LO_2$ produced that is delivered to LEO
Ground support manpower	$n_b$	Number of full-time equivalent heads
Ground support overhead factor	$n_f$	Multiplier of manpower needed for total costs

man-year of ground support manpower. The variable that precedes this constant,  $n_f$ , is a ground support overhead factor that is multiplied by manpower costs to obtain total ground support costs.

After setting up these costs equations, baseline values were assigned to each cost variable, with the ground rule that lowest risk technologies would be utilized for each system. Lunar base modules, for example, were assumed to be modified versions of the habitat, laboratory, and logistics modules that will be developed for NASA's LEO space station.

Another key ground rule was that selection of the lunar processing site would be made on the basis of scientific data whose costs would not be included in this model. It was further assumed that an initial lunar base would be in place prior to the  $LO_2$  production activity and that this facility would be scaled up to meet the  $LO_2$  production requirements. The costs included in this model are only the marginal costs of expanding this initial facility to meet  $LO_2$  production objectives.

Although some of these ground rules resulted in lower capital and operations cost estimates, the specification of lowest risk technology prevented the incorporation of many potentially cost-reducing technologies into the baseline scenario. The probable impact of these ground rules is analyzed later in this paper.

## RESULTS OF THE ANALYSIS

Once baseline values were assigned to the cost variables, a simple calculation was made to obtain baseline capital and operations cost estimates. These costs were determined to be \$3.1 billion for baseline capital cost and \$885 million/per year for baseline operations costs.

An analysis of the performance of lunar OTVs indicated that 49.2% of the LO<sub>2</sub> produced would be delivered to LEO. Consequently, the unit cost of LO<sub>2</sub> delivered to LEO, assuming 10-year amortization of capital costs, was determined to be \$2,430/kg (\$1,100/lb). This cost is about one third less than the current cost of utilizing the STS for propellant delivery to LEO and roughly equivalent to STS cost projections in the most optimistic, high flight-rate, low operations cost scenario. It should be reemphasized, however, that these costs are based on a specific set of assumptions and are for comparative purposes only.

The most important objectives of this analysis were the assignment of uncertainty ranges to each of the cost variables, the calculation of the sensitivity of LO<sub>2</sub> production costs to each of these variables, and the analysis of the technical and programmatic assumptions used to arrive at values for each variable. The data that were developed to support the sensitivity analysis of space resource utilization cost variables are summarized in Table 2. The baseline, best case, and worst case values assigned to each cost variable are shown, along with the impact of each variable's best case and worst case values on capital and operations costs. For example, as power requirements vary from a low value of 4 Mw to a high value of 12 Mw, with all other variables held at their baseline values, the capital cost for establishing the baseline capability ranges from \$2.30 billion to \$3.90 billion.

From this table it is evident that the principal driver of capital costs is the lunar base power requirement, while the Earth-to-Moon transportation cost is the most important operations cost driver. Since capital costs are amortized over a 10-year period, the Earth-to-Moon transportation cost has a much greater overall impact on the cost of lunar LO<sub>2</sub> in LEO. If this cost could be reduced from its baseline value of \$10,000/kg delivered to the Moon to its best case value of \$5,000/kg, capital costs would drop from \$3.1 billion to \$2.45 billion, operations costs would decline from \$885 million/yr to \$468 million/yr, and the cost of lunar LO<sub>2</sub> would be reduced from \$2,430/kg to \$1,450/kg. Conversely, at its worst case value of \$15,000/kg, the Earth-to-Moon transportation cost would drive capital costs up to \$3.75 billion, operations costs to \$1.3 billion/yr, and the cost of lunar LO<sub>2</sub> to \$3,410/kg.

An alternative approach to showing the sensitivities of the cost variables is illustrated in Table 3, where percent changes in capital costs, operations costs, and the costs per kilogram of LO<sub>2</sub> produced (with 10-year amortization of capital costs) are shown. In this table the variables are ranked and listed in order of their impact on the LO<sub>2</sub> cost/kg. The influence of each variable is calculated as an "impact factor" equal to the average of the best case and worst case percent changes in LO<sub>2</sub> cost/kg due to extreme values of the variable. From these impact factors it is clear that two of the cost factors are

Table 2. Capital and Operations Costs—Sensitivity to Key Variables

	Baseline Case	Best Case		Worst Case	
	Most Likely Value	Value	Result	Value	Result
<i>Capital Costs</i>					
1. Power required	8 Mw	4 Mw	\$2.30 B	12 Mw	\$3.90 B
2. Cost of power	\$100 M/Mw	\$50 M/Mw	\$2.70 B	\$200 M/Mw	\$3.90 B
3. Number of types of lunar base modules	1	0	\$2.80 B	2	\$3.40 B
4. Cost of modifying space station modules	\$300 M	\$100 M	\$2.90 B	\$500 M	\$3.30 B
5. Number of lunar base modules	1	1	\$3.10 B	3	\$3.90 B
6. Unit cost of lunar base modules	\$200 M	\$100 M	\$3.00 B	\$300 M	\$3.20 B
7. Processing/storage facility cost	\$500 M	\$300 M	\$2.90 B	\$1.0 B	\$3.60 B
8. Power system mass	10,000 kg/Mw	5,000 kg/Mw	\$2.70 B	15,000 kg/Mw	\$3.50 B
9. Earth-to-Moon transportation cost	10,000/kg	5,000/kg	\$2.45 B	15,000/kg	\$3.75 B
10. Mass of lunar base modules	20,000 kg	15,000 kg	\$3.05 B	30,000 kg	\$3.20 B
11. Mass of processing/storage facility	30,000 kg	15,000 kg	\$2.95 B	50,000 kg	\$3.30 B
<i>Operations Costs</i>					
1. Number of lunar base resupply missions/yr	1	1	\$885 M/yr	3	\$1.285 B/yr
2. Net lunar oxygen delivered to LEO	49.2%	70%	\$625 M/yr	30%	\$1.125 B/yr
3. Ground support manpower	20	10	\$860 M/yr	50	\$960 M/yr
4. Ground support overhead factor	25	5	\$845 M/yr	50	\$935 M/yr
5. Earth-to-Moon transportation cost	\$10,000/kg	\$5,000/kg	\$468 M/yr	15,000/kg	\$1.303 B/yr
6. Mass of lunar base modules	20,000 kg	15,000/kg	\$835 M/yr	30,000 kg	\$985 M/yr

Table 3. Sensitivity of Capital, Operations, and Oxygen Production Costs to Uncertainty Ranges of Key Variables

Variable	Sensitivity Ranking	Best Case		Worst Case		Impact Factor
		Change in Capital Cost	Change in LO <sub>2</sub> Cost/kg	Change in Capital Cost	Change in LO <sub>2</sub> Co st/kg	
<i>Capital Costs</i>						
Earth-to-Moon transportation cost	1	-21%	-40%*	+21%	+40%	40
Power required	2	-26%	- 7%	+26%	+ 7%	7
Mass of lunar base modules	3	- 2%	- 4%	+ 3%	+ 9%*	7
Cost of Power	4	-13%	- 3%	+26%	+ 7%	5
Number of lunar base modules	5	0%	0%	+26%	+ 7%	4
Power System mass	6	-13%	- 3%	+13%	+ 3%	3
Processing/storage facility cost	7	- 6%	- 2%	+16%	+ 4%	3
Number of types of lunar base modules	8	-10%	- 3%	+10%	+ 3%	3
Cost of modifying space station modules	9	- 6%	- 2%	+ 6%	+ 2%	2
Mass of processing/storage facility	10	- 5%	- 1%	+ 6%	+ 2%	2
Unit cost of lunar base modules	11	- 3%	- 1	+ 3%	+ 1%	1
<i>Operations Costs</i>						
Net lunar oxygen delivered to LEO	1	-29%	-45%	+27%	+97%	71
Earth-to-Moon transportation cost	2	-47%	-40%*	+47%	+40%*	40
Number of lunar base resupply missions/yr	3	0%	0%	+45%	+13%	7
Mass of lunar base modules	4	- 6%	- 4%*	+11%	+ 9%*	7
Ground support manpower	5	- 3%	- 3%	+8%	+ 6%	5
Ground support overhead factor	6	- 5%	- 3%	+6%	+ 4%	4

\*Impact based on changes in both capital costs and operations costs.



by far more important than all the rest: Earth-to-Moon transportation costs and net delivery of  $\text{LO}_2$  to LEO. The net percentage of  $\text{LO}_2$  delivered to LEO is important because of its double impact. As the percentage of  $\text{LO}_2$  delivered declines,  $\text{LO}_2$  cost/kg increases not only because less  $\text{LO}_2$  is delivered, but also because more hydrogen must be provided from the Earth to augment the  $\text{LO}_2$  used as propellant from the Moon to LEO.

Table 3 also shows that costs associated with establishing crew accommodations on the Moon, e.g., the number and cost of lunar base modules, have a relatively small impact on total cost. Lunar base resupply requirements, however, are a major driver, as is the mass of lunar base modules. Factors influencing the mass and cost of the power system and the processing/storage facility have a significant impact on capital costs but have a much smaller effect on  $\text{LO}_2$  cost/kg. Factors affecting ground support costs have a moderate impact on operations costs and  $\text{LO}_2$  cost/kg.

It is not surprising that the six operations cost variables are among the nine most important cost factors. Operations costs are significant because they are driven by the high cost of Earth-to-Moon transportation, while the impact of capital costs is reduced because they are amortized over a 10-year period. The relative significance of the operations cost leads to the important observation that  $\text{LO}_2$  production costs may be reduced substantially by increasing capital expenditures on technologies that can reduce operations costs.

## INTERPRETATION OF ANALYSIS RESULTS

During this study, a more detailed sensitivity analysis was performed to obtain a preliminary indication of which technologies need to be developed to maximize the probability that lunar  $\text{LO}_2$  production will become a cost-effective enterprise. It became evident that the cost-effective performance of such an operation will depend primarily on the achievement of three major cost-reducing objectives: (1) reducing or eliminating the need to transport hydrogen from Earth to the Moon; (2) reducing space transportation costs, particularly the cost of Earth-to-Moon transportation; and (3) reducing lunar base resupply requirements.

If all three of these objectives were met to the greatest extent possible, *i.e.*, if hydrogen transportation requirements were eliminated, Earth-to-Moon transportation costs were reduced to its best-case value, and lunar base resupply requirements were eliminated, the cost/kg of lunar  $\text{LO}_2$  delivered to LEO would be reduced from \$2,340/kg to \$600/kg, or about \$270/lb. These figures assume no change in capital costs; but even if capital costs were doubled to achieve these capabilities,  $\text{LO}_2$  cost/kg would be reduced to approximately \$1,100/kg, less than half the baseline cost.

Twenty-five key technology issues influencing these and the other  $\text{LO}_2$  production cost factors are presented in Table 4. In this table, the impact of each technology issue on the 15 cost variables is shown; a darkened square indicates a strong impact; a light square represents a moderate effect; no square indicates little or no impact. It should be emphasized that the selection and evaluation of these technology issues was based on the subjective judgment of a panel of experts convened to support this study, rather

Table 4. Impact of 25 Key Technology Issues on Space Resource Utilization Cost Factors

	Net Lunar LO <sub>2</sub> delivered to LEO	Earth-to-Moon transportation cost	Mass of lunar resupply modules	Power required	Cost of power	Ground support modules	Ground support missions/yr	Number of lunar base modules	Processing overhead factor	Mass of processing facility	No. types of processing facility	Cost of processing facility	Unit cost lunar base modules	Cost modifying space station modules	Unit cost lunar base modules	Unit cost lunar base modules
<div> <div></div> heavy impact         </div> <div> <div></div> moderate impact         </div> <div> <div></div> little or no impact         </div>																
Lunar base power source (nuclear vs. solar)																
Scalability of small (<100 Kw) power systems																
Electrical vs. thermal energy																
Power consumption of processing technique(s)																
Complexity of power system installation																
Maintainability of power system																
Pressurized volume required for lunar operations																
Duration of lunar base crew shifts																
Degree of automation of lunar base operations																
Lunar base module commonality w/space station nodules																
Lunar base shielding requirements																
Space station interfaces																
Scalability of initial lunar research facilities																
Complexity of lunar factory processes																
Number of lunar factory processes																
Commonality of processing facility w/space station lab modules																
Commonality of LO <sub>2</sub> storage unit w/OTV propellant depot																
Performance and cost of SDLV/HLLV (if available)																
Performance and cost of OTVs (if available)																
Size of lunar base crew																
Degree of closure of lunar base life support system																
Availability of aerobrake for LO <sub>2</sub> delivery																
Availability of lunar hydrogen																
Self-sufficiency of lunar operations																
Ground support philosophy																

than a quantitative analysis. Based on the sensitivity analyses presented in this paper, the 15 cost factors are listed along the top of Table 4 in descending order of importance from left to right. Hence, a visual scan of Table 4 provides an immediate glimpse of the relative importance of the technologies based on three considerations: total number of squares, number of dark squares, and distribution of squares to the left of the chart (*i.e.*, toward the most important cost factors).

To quantify the impact of these 25 technology issues on the economics of the baseline space resource utilization scenario, a technology weighting factor of 3 was assigned to each dark square and 1 to each light square. These technology weighting factors then were multiplied by the impact factor (see Table 3) for each cost factor that the technology issue affects. A total economic weighting factor was calculated for each technology issue, based on the sum of the products across each row. For example, the lunar base power source has a heavy impact on power system mass and cost of power, for a total economic weighting factor of  $(3 \times 5) + (3 \times 5) = 30$ .

The 10 most important technology issues, based on their total economic weighting factors, are listed in Table 5. All 10 issues relate in some way to space transportation costs and/or lunar base resupply requirements, with performance and cost of space-based orbital transfer vehicles (OTVs) being the most critical technology issue. Developing a low cost OTV is a fundamental requirement for cost-effective space resource utilization because the OTV is the single most effective means of reducing Earth-to-Moon transportation costs.

The second most important issue is the availability of lunar hydrogen, since production of hydrogen on the Moon currently appears to be the most promising solution to the problem of costly hydrogen deliveries from Earth. The technology issue ranked third is availability of an OTV aerobrake; the capability for aeroassisted return to LEO is a requirement for achieving the efficiency in  $\text{LO}_2$  delivered to LEO, thereby halving the effective  $\text{LO}_2$  cost/kg. The fourth most important technology issue is the performance and cost of shuttle-derived launch vehicles (SDLVs) or heavy-lift launch vehicles (HLLVs), *i.e.*, the Earth-to-LEO transportation cost. However, it should be pointed out that cost reductions in this area will also reduce the cost of delivering  $\text{LO}_2$  from Earth to LEO, making it more difficult for the lunar  $\text{LO}_2$  production scheme to compete.

Table 5. Major Technology Issues

1. Performance and cost of OTVs (345)
2. Availability of lunar hydrogen (254)
3. Availability of aerobrake for $\text{LO}_2$ delivery (213)
4. Performance and cost of SDLV/HLLV (120)
5. Degree of automation of lunar base operations (119)
6. Self-sufficiency of lunar operations (97)
7. Size of lunar base crew (79)
8. Degree of closure of lunar base life-support system (71)
9. Complexity of lunar factory processes (51)
10. Number of lunar factory processes (48)

Numbers in parentheses are economic weighting factors.

The remaining six of the “top ten” technology issues all pertain to lunar base resupply requirements. The extent of lunar base automation and self-sufficiency are the most important of these issues, with the next two factors relating to the resupply needs of the lunar base crew. The ninth and tenth most significant technology issues relate to the complexity and number of lunar factory processes.

## CONCLUSIONS AND RECOMMENDATIONS

On the basis of the analysis presented in this paper, it is not possible to establish conclusively whether production of liquid oxygen from lunar materials can be justified on economic grounds. Although the cost estimates for the baseline scenario are encouraging, a number of technologies with significant impact on lunar LO<sub>2</sub> production costs must be explored.

The principal conclusion is that the costs of any space resource utilization venture are likely to be dominated by transportation costs. This situation may change when the focus of human space activities moves sufficiently far from Earth (e.g., once large scale space industrialization is underway) but will probably remain valid as long as space resources are developed for use in near-Earth space. For this reason, it is recommended that high priority be given to development and evaluation of more cost-effective space transportation systems, particularly low-cost OTVs with an aeroassisted return-to-LEO capability.

Another key issue relating the baseline scenario is the cost of Earth-derived hydrogen. Production of lunar LO<sub>2</sub> would be far more cost-effective if a capability for the co-production of lunar hydrogen could be developed, even if capital costs were increased substantially. Although relatively large quantities of lunar ore would need to be processed, the additional costs of lunar hydrogen production could be offset by savings of over \$600 million/yr in transportation costs. Production of some alternative propellant constituent, such as aluminum, also might offer an opportunity for reducing or eliminating costly imports of fuels from Earth. However, this latter approach would require an entirely new development, the design of aluminum-burning space engines.

A third category that seems to have a substantial impact on the economics of lunar resource utilization are the technologies influencing lunar base resupply requirements. Increasing lunar base automation, closing the lunar base life support system, and other steps to reduce the frequency and scale of resupply missions appear to have a high likelihood of providing economic benefits and should be given particular emphasis in future study efforts.

Finally, it is important that parametric cost analyses be used in the assessment of a variety of space resource utilization scenarios. Use of lunar ore for production of construction materials is one such option, although this type of enterprise would probably require a dramatic increase in the level of space activity to be cost effective. Another option that merits careful consideration is the development of asteroidal resources. Both rocket propellants and construction materials could be derived from asteroids, and while the up-front costs of asteroid utilization would probably exceed the capital expenditures

required for lunar development, operations costs could be substantially lower. Further analysis of all these opportunities needs to be conducted over the next several years before a commitment is made to any particular plan for space resource utilization.

As these and related technologies are developed, the reliability of space resource utilization cost estimates will improve. Eventually, it will become possible to generate cost estimates of sufficient fidelity to support detailed definition of space resource utilization objectives. An important step in this process will be the adaptation of this parametric model and similar techniques to the evaluation of a broad range of space resource development options.

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# LUNAR OXYGEN PRODUCTION FROM ILMENITE

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Lunar oxygen production from ilmenite appears to be the simplest means available. Fixed- or fluidized-bed, gas-solid reactor processing systems appear suitable for development; terrestrial analogs exist. Scale of operations for 1000 metric tonnes per year seems feasible. Severity of the lunar environment calls for particular design attention to fluid-solid flow, heat exchange, power supply, minimum maintenance, and modular replacement.

## INTRODUCTION

In any future human colonization of the Moon, oxygen is clearly one of the most important materials to be supplied. It is required for both life support and propulsion. Incentives for lunar oxygen production from lunar raw materials as opposed to supplying it solely from Earth have been discussed by Criswell (1983) and Davis (1983). Both conclude that more efficient use of Earth-supplied hydrogen, total propellants, and available payload weights and volumes all result from lunar oxygen production.

Two types of lunar materials have been proposed as raw materials for oxygen production: ilmenite,  $\text{FeTiO}_3$ , and silicates such as anorthite,  $\text{CaAl}_2\text{Si}_2\text{O}_8$  (Williams and Erstfeld, 1979; Kesterke, 1971; Steurer, 1982; Carroll, 1983). Both are lunar-surface-minable, occurring in soils, breccias, rocks, and basalts. The silicates are considerably more abundant than ilmenite, and this would suggest a preference for them as source materials. However, the silicates present more difficult process engineering problems because they must be reduced at temperatures of 1100°C or more by fluxed molten-phase electrolysis (Kesterke, 1971). Alternatively, silicates can be processed at still higher temperatures by plasma processes or, if flux is not used, in electrolysis processes (Steurer, 1982; Carroll, 1983). Under these conditions, degradation of electrode and/or container materials and difficult recovery of Earth-imported electrolyte fluxing agents present severe challenges that are, at present, difficult to overcome. Ilmenite, on the other hand, can be reduced at temperatures of 1000°C or below, and it can be separated and concentrated from a natural abundance of approximately 5 wt % to perhaps 90 wt % in the process feedstock (Williams *et al.*, 1979). Williams *et al.* (1979) have proposed electrostatic and magnetic schemes for beneficiation; Agosto (1984) has performed initial and encouraging experimental tests of one of these beneficiation schemes. These circumstances make ilmenite appear to be the preferable raw material.

The available processing techniques include hydrogen-reduction, chemical reductions with other reagents, and electrochemical reduction (Rao *et al.*, 1979). We have concluded that probably the best route to lunar oxygen is via hydrogen-reduction of ilmenite. This is best done with some kind of continuous or semi-continuous process cycle using recycled,

pressurized hydrogen as the working fluid. This paper discusses the various reduction options along with the reasons for our design choices.

## ILMENITE REDUCTION

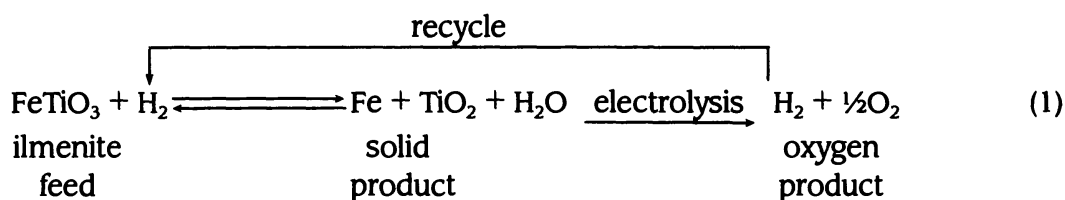
### Choice of Reductant

Hydrogen was chosen over several other potential reducing agents or methods, e.g.,  $\text{CH}_4$ , CO, and electrolysis, for the following reasons:

- Hydrogen will probably have to be imported for propulsion and life support anyway, so it is likely to be the easiest material to supply for make-up of processing losses caused by leakage from the recycle loop.
- With carbon-bearing reductants ( $\text{CH}_4$  and CO), part or all of the oxygen product must be won by electrolyzing mixtures containing CO and  $\text{CO}_2$ . This step often leads to solid carbon deposits that are hard to remove continuously. With  $\text{H}_2$ , the electrolysis of liquid  $\text{H}_2\text{O}$ , at least, is straightforward, and this one step liberates the oxygen product and regenerates the reducing agent.
- As discussed below, use of  $\text{H}_2$  may present some heat integration opportunities with large energy-saving consequences. These would not be possible with an electrochemical or  $\text{CH}_4$ -reduction technique, neither of which allow regenerating the reducing agent at reduction temperature.
- Electrochemical methods require solutions to the difficult container and electrode materials problems mentioned above.

### Process Scheme

The basic reactions are shown below.



The process requires heating and contacting of gas and solids, and, to be fully continuous, a way of introducing solid ilmenite feed and removing spent, reduced ilmenite product must be provided. It would be very desirable to use countercurrent, gas-solids heat exchange to reduce overall energy requirements and to reject heat in spent solids at as low a temperature as possible. It would also be desirable to conduct the  $\text{H}_2\text{O}$  electrolysis at reaction temperature (vapor phase electrolysis), if possible, to avoid cooling and reheating the recycled  $\text{H}_2$  to condense  $\text{H}_2\text{O}$ .

Figure 1 presents a conceptual design of a fluidized bed reactor/vapor phase electrolysis flow plan that at least on paper accomplishes the objectives above. The reactor

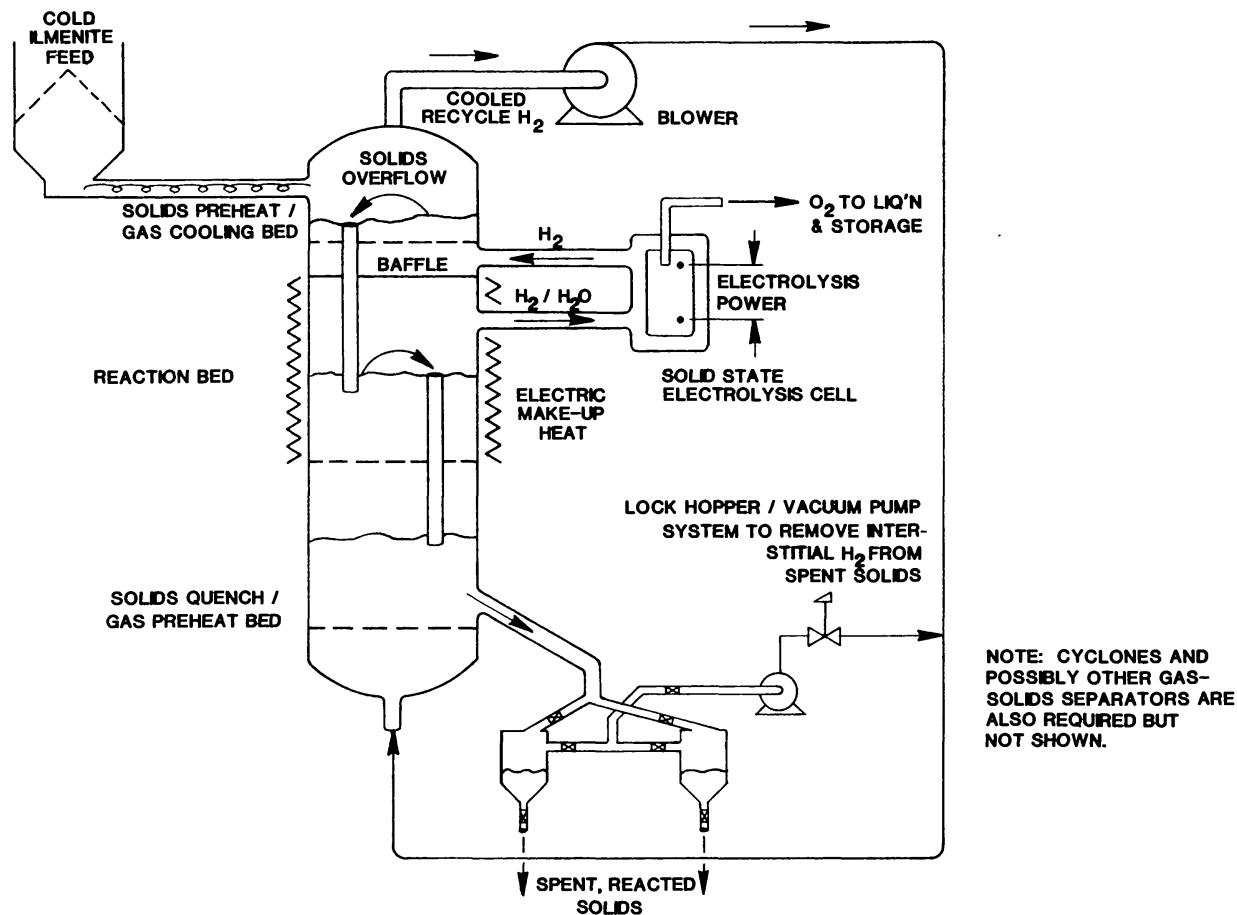


Figure 1. Continuous, fluid bed ilmenite reduction/ $O_2$  production.

shell actually contains three staged fluidized beds stacked vertically with gas in upward flow and solids in countercurrent downward flow. Cold ilmenite feed from a lock hopper at reactor pressure enters the top, solids-preheat bed via a screw feeder. Here it exchanges heat with hot recycled  $H_2$ . This stage preheats the solids and cools the  $H_2$  sufficiently for it to be circulated reliably by the blower. The solids overflow into the reaction bed where the  $H_2$  reduction occurs. Electrical make-up heat must be supplied to this stage on temperature control to bring gas and solids to the desired reaction temperature and to supply the endothermic heat of reaction.

Finally, spent, reacted solids overflow into the solids' quench/gas-preheat bed where they exchange heat with cold, recycled  $H_2$ . The cooled solids are discharged through an alternating lock hopper/vacuum pump apparatus to minimize loss of interstitial  $H_2$  gas to the lunar vacuum.

To extract product oxygen, the  $H_2/H_2O$  mixture leaving the reaction bed is sent to an electrolyzer using a solid-state, ceramic electrolyte and operating at reactor temperature. This device was suggested by the prototype studies of Weissbart and Ruka (1962). Gaseous oxygen is evolved on the product side of the solid electrolyte and is sent to liquefaction



and storage. The  $\text{H}_2\text{O}$  is reconverted to  $\text{H}_2$  on the feed side of the electrolyte, and the  $\text{H}_2$  is recycled.

Staged, fluidized-bed reactor/heat exchangers such as in Fig. 1 are in commercial use for terrestrial applications such as limestone calcining and sulfide roasting (Perry and Chilton, 1973). Their use was also proposed for fluidized iron ore reduction (FIOR). This was a process developed and studied intensively in the 1960s and 1970s by Exxon Corporation and several steel companies. Iron ores were reduced in fluidized beds with  $\text{H}_2$ , natural gas, and other reducing gases as a possible replacement for conventional blast furnaces. Changing energy economics prevented exploitation of the process, but its strong analogies to  $\text{H}_2$ -reduction of ilmenite are evident.

As noted in Fig. 1, efficient cyclones and possibly other types of gas-solids separators will be required but, for clarity, are not shown. Clean, gas-solid separations will be especially important upstream of the blower and the solid-state electrolysis device. Proper sizing of feed solids and rejection of fines will also alleviate entrainment and elutriation problems.

In addition to heat integration, the scheme of Fig. 1, if workable, would have significant heat transfer equipment advantages. All heating of gases and solids, except for  $\text{O}_2$  cooling and liquefaction, occurs in fluidized beds with very high transfer coefficients. In particular, if the solid-state, high-temperature electrolysis proved feasible, the poor-coefficient process of condensing dilute  $\text{H}_2\text{O}$  vapor from a "fixed" gas,  $\text{H}_2$ , would be avoided.

### Thermodynamic Considerations

The ilmenite reduction step of (1) is mildly endothermic;  $\Delta H = +9.7$  kcal/gm-mol at  $900^\circ\text{C}$  as calculated from Williams and Erstfeld's data (1979). Reduction is, however, reversible and strongly equilibrium-limited. Figure 2 shows equilibrium conversion of a pure  $\text{H}_2$  feed as a function of temperature. At  $900^\circ\text{C}$ , the per-pass conversion of  $\text{H}_2$  cannot exceed 7.4%. This is important because it means that required  $\text{H}_2$  circulation rates are large compared to  $\text{O}_2$  production rates since

$$\text{Required molar } \text{H}_2 \text{ circulation rate} = \frac{2 \cdot \text{Required molar } \text{O}_2 \text{ production rate}}{(\text{H}_2 \text{ conversion})} \quad (2)$$

Large circulation rates imply substantial premiums on devising energy-efficient ways to circulate the recycled  $\text{H}_2$  with minimal temperature cycling. Both the countercurrent gas/solids heat exchange and the solid-state electrolyzer of Fig. 1 contribute to this type of improved energy efficiency.

### Scale of Operations

Initial lunar base planning indicates the need for an oxygen production facility producing approximately 1000 metric tonnes of liquid  $\text{O}_2$  product annually. Reasonable assumptions of reactor operating conditions are as follows: temperature =  $900^\circ\text{C}$ ; per-pass hydrogen conversion = 5% (about 2/3 of the equilibrium value); superficial gas velocity in fluid

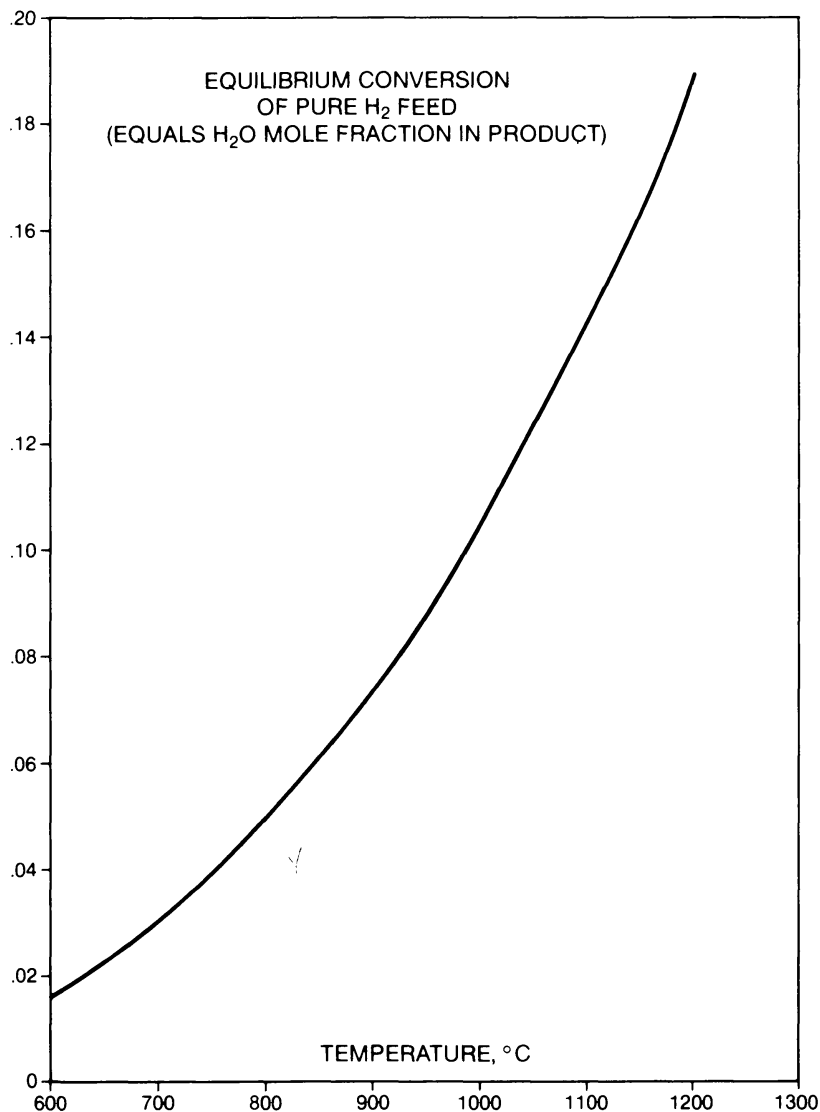


Figure 2. Equilibrium for the reaction  $\text{FeTiO}_3 + \text{H}_2 \rightleftharpoons \text{Fe} + \text{TiO}_2 + \text{H}_2\text{O}$ .

beds = 1 ft/s; gas velocity in circulating line = 50 ft/s. These assumptions lead to an estimated hydrogen circulation rate of about 2200 standard cubic feet per minute, about the capacity of a 14- to 16-inch diameter ventilating fan. The reactor and circulation line diameters can be estimated as a function of operating pressure as shown in Table 1. Note that these assumptions fix only the reactor *diameter* and not the bed heights or volumes. Specifying heights or volumes requires reaction rate, heat transfer, and fluidized bed expansion data. We are presently obtaining these data in our experimental and engineering study.

These sizes appear to pose no problem, particularly at the higher pressures. Engineering evaluations are necessary to find the best balance between smaller diameter and thicker walls at high pressures versus larger diameters but thinner walls at low pressures.

Table 1. Reactor and Circulating Line Diameters

Operating Pressure, KPa	Reactor Diameter	H <sub>2</sub> Circulation Line Diameter
100 (1 atm)	13.5'	12"
200 (2 atm)	9.5'	10"
500 (5 atm)	6.0'	6"
1000 (10 atm)	4.5'	4"

### Other Design Considerations Specific to Lunar Operations

Tables 2–4 list lunar environmental features differing greatly from Earth and identify specific consequences of these features on the design. Tables 2 and 3 discuss the more familiar, broad implications applying to design of any lunar processing plant. Table 3, in particular, shows design responses required to minimize adverse effects on plant operability/service factors. Table 4 concentrates on reduced gravity effects and shows the quantitative effect of gravitational acceleration on several important fluidized bed parameters. These results were derived from general fluidization correlations collected in Davidson and Harrison (1963) and Kunii and Levenspiel (1969). The overall conclusion from Table 4 is that, compared to terrestrial fluid beds with similar gas rates, lunar fluidized

Table 2. Lunar Versus Terrestrial Environment—Effects on Processing

Environmental Feature	Comparison to Earth	Processing Consequences
Gravity	Moon: 1/6 g Earth: 1 g	Major effects on fluidized beds, gas-solids transport systems, gravity flow of liquid and particulate solids
Surface Temp. Range	Moon: About 290°C (-140°C–+150°C) Earth: 30°C	Widely fluctuating as-mined feed-solids temperature
Atmosphere/ Coolants	Moon: Air/Water Absent Earth: Air/Water Abundant	Only closed-loop fluid systems usable; final heat rejection by radiation or heat pipe; unlimited hard vacuum available
Conventional Fuels	Moon: Absent Earth: Plentiful	Process heating by electricity or direct solar; power generation by nuclear or solar
Human Access	Moon: Difficult/Minimal Earth: Easy/Frequent	Extreme emphasis on minimum maintenance, modular replacement

Table 3. Other Lunar Environment/Design Effects

Environmental Feature	Design Response
Fluctuating Surface (Feed Solids) Temperature	Provide agitated holding bins to average out Overdesign preheat capacity
Lack of Coolants/Conventional Fuels	Use heat integration to reduce energy demand, heat rejection duties  High heater-to-process coefficients desirable for make-up heat supply Efficient, low-weight radiators desirable
Difficult Human Access	Redundancy/automated change-out for high-maintenance items: Pumps and blowers Solids feeders Electric resistance heaters Overdesign/minimize use of high-wear items Shaft seals Rotating surfaces in dusty or gritty service

beds will need larger particles and/or lower fluidizing velocities and will expand substantially more. The larger particles may not have the adverse effect of larger bubbles/poorer contacting efficiency that they would have on Earth.

Table 4. Reduced Gravity Effects on Equipment Design

Parameter	Approximate Dependence on g	Lunar vs. Terrestrial Design
<i>Fluidized Bed Reactors, Solids Standpipes</i>		
Minimum Fluidization Velocity, $U_{mf}$	$g^{1.0}$	Operable gas velocity range is from $U_{mf}$ to $U_t$ ; must use larger particles or lower velocities
Particle Terminal Velocity, $U_t$	$g^{2/3}$ – $g^{1.0}$	Larger particles→larger bubbles→poorer contacting efficiency
Bubble Diameter	$g^{0.4-1.0}$	Smaller bubbles mean better contacting efficiency; gravity effect counters particle size effect on bubble size
Bed Expansion	$1/(g^{0.7-1.0})$	Taller bed required for same inventory
Standpipe Throughput	$g^{0.5}$	Taller standpipes for same throughput
<i>Fixed Bed Reactors</i>		
No major effects		
<i>Liquid Pumps</i>		
Suction Head	$g^{1.0}$	Taller suction legs or low NPSH pumps required

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# **OXYGEN EXTRACTION FROM LUNAR MATERIALS: AN EXPERIMENTAL TEST OF AN ILMENITE REDUCTION PROCESS**

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The reaction of ilmenite with hydrogen to produce water has been studied experimentally in order to evaluate the effectiveness of using a cold trap to improve yields in a continuous flow process. Yields were enhanced, but not to the degree observed in batch processing systems. The terrestrial simulant used in these studies contained traces of iron sulfide, which released H<sub>2</sub>S during processing with a deleterious effect on several components of the test system. More sophisticated testing should be undertaken to obtain kinetic data and attention given to the removal of sulfides in the pre-process beneficiation.

## **INTRODUCTION**

Several studies (e.g., Driggers, 1976; Bock, 1979) have indicated that large quantities of propellant are required to support orbital transfer operations in an expanded space transportation system. A large energy penalty is paid for lifting propellant from the Earth's surface to orbits at which it is required. If a source for propellant were available in space, or at least from sources with a shallower gravity well than the Earth's, the efficiency of an orbital transfer system could be improved (Criswell, 1983; Davis, 1983). Other studies (e.g., Rao *et al.*, 1979; Criswell, 1978, 1980) have proposed that the rocks and minerals found on the Moon are a source of oxygen for use as the oxidizer in H<sub>2</sub>/O<sub>2</sub> propulsion systems. The reduced energy cost associated with lifting the oxygen into space from the Moon's relatively shallow gravity well, combined with the fact that oxygen makes up 86% by weight of propellant in LOX-H<sub>2</sub> systems, suggests that substantial net benefit could accrue to the overall transportation system if the production of LOX from lunar materials is feasible.

One of the most simple of the possible extraction processes involves the reaction of hydrogen with iron-oxide-bearing lunar minerals to produce water, from which hydrogen and oxygen are then extracted by electrolysis. Ilmenite (FeTiO<sub>3</sub>) is an ideal lunar mineral on which to base an extraction process: it is the most abundant of the oxide minerals (up to 10%, modal, in some Apollo 11 soils); it can be readily separated from other non-reactive soil components (Agosto, 1983); it is relatively rich (58% by weight) in the reducible FeO component; and it is chemically quite pure, so that the possibility of side reactions is reduced. In addition, the production reaction,  $\text{H}_2 + \text{FeTiO}_3 = \text{Fe} + \text{TiO}_2 + \text{H}_2\text{O}$ , is very similar (chemically and thermodynamically) to those involving other iron oxides and iron silicates; thus, a study of it serves as a good general model. The major technical problem

is that at moderate temperatures (700–1000°C) the per pass conversion of  $H_2$  to  $H_2O$  is less than 5%.

Although the conversion can be enhanced by running the process at higher temperatures, major problems (e.g., degradation of materials, hydrogen loss by diffusion, and sintering of solid reactants and products) occur that limit the viability of this solution. Williams *et al.* (1979) suggest that the per pass yield could be substantially improved by buffering the  $H_2O$  pressure over the reaction at the water liquid-vapor equilibria. Essentially, a cold trap is included in the system; water is pumped from the reaction zone producing a greatly enhanced yield. Williams and Mullins (1983) reported preliminary experimental results that verified that the process modification by Williams worked in a batch mode. It is the purpose of this report to present the results of a study of a variation of the process suitable for operation in a continuous mode.

## THEORY

If the solid phases are pure and if the pressures and temperatures are such that the gases behave ideally, the reaction  $H_2 + FeTiO_3 = Fe + TiO_2 + H_2O$  is described by the equation

$$K = PH_2/PH_2O \quad (1)$$

in which  $K$  is function of temperature only and is computable from basic thermochemical data, and  $PH_2$  and  $PH_2O$  are the equilibrium partial pressures of hydrogen and water. Defining  $P_{lv}$  to be the pressure of  $H_2O$  in equilibrium with liquid and constraining  $PH_2O = P_{lv}$ , (1) becomes

$$K = PH_2/P_{lv}$$

Since  $P_{lv}$  is primarily a function of the cold trap temperature, it will not be the same as  $PH_2O$  (equation 1) except fortuitously; thus,  $PH_2$  must be different from that in (1). We will call it  $PH_2'$  and write

$$K = PH_2'/P_{lv} \quad (2)$$

The per pass conversion of  $H_2$  is defined as

$$C = 1 - (P - P^iH_2)/P^iH_2$$

where  $P$  is either the hydrogen pressure given by (1) or (2), and  $i$  denotes the initial hydrogen pressure. The calculated conversions are summarized in Table 1. Comparing the conversions, one sees that the incorporation of a cold trap theoretically can improve yields by as much as 23-fold. Of course, the calculations assume thermodynamic equilibrium. Williams and Mullins (1983) reported on experiments that demonstrated that in the batch mode (that is, the one in which overall equilibrium should be attained)

Table 1. Theoretical Yields

T(°C)	C(W/O Cold Trap)	C(W/Cold Trap)
600	0.031	0.73
700	0.044	0.81
800	0.057	0.85

Notes: C (W/O Cold Trap) computed from Equation 1.

C (W/O Cold Trap) from Equation 2, using an initial hydrogen pressure at 10 psi and cold trap temperature of 10°C.

Thermodynamic data from Robie *et al.* (1978).

enhancements in yield were obtained using "cold-trap technology" that were consistent with theory. Here we discuss follow-on experiments in which recirculation of hydrogen was simulated; in this type of process, equilibrium will only be *approached* due to flows in the system. The major issue here is whether any enhancement in yield is produced using a cold trap.

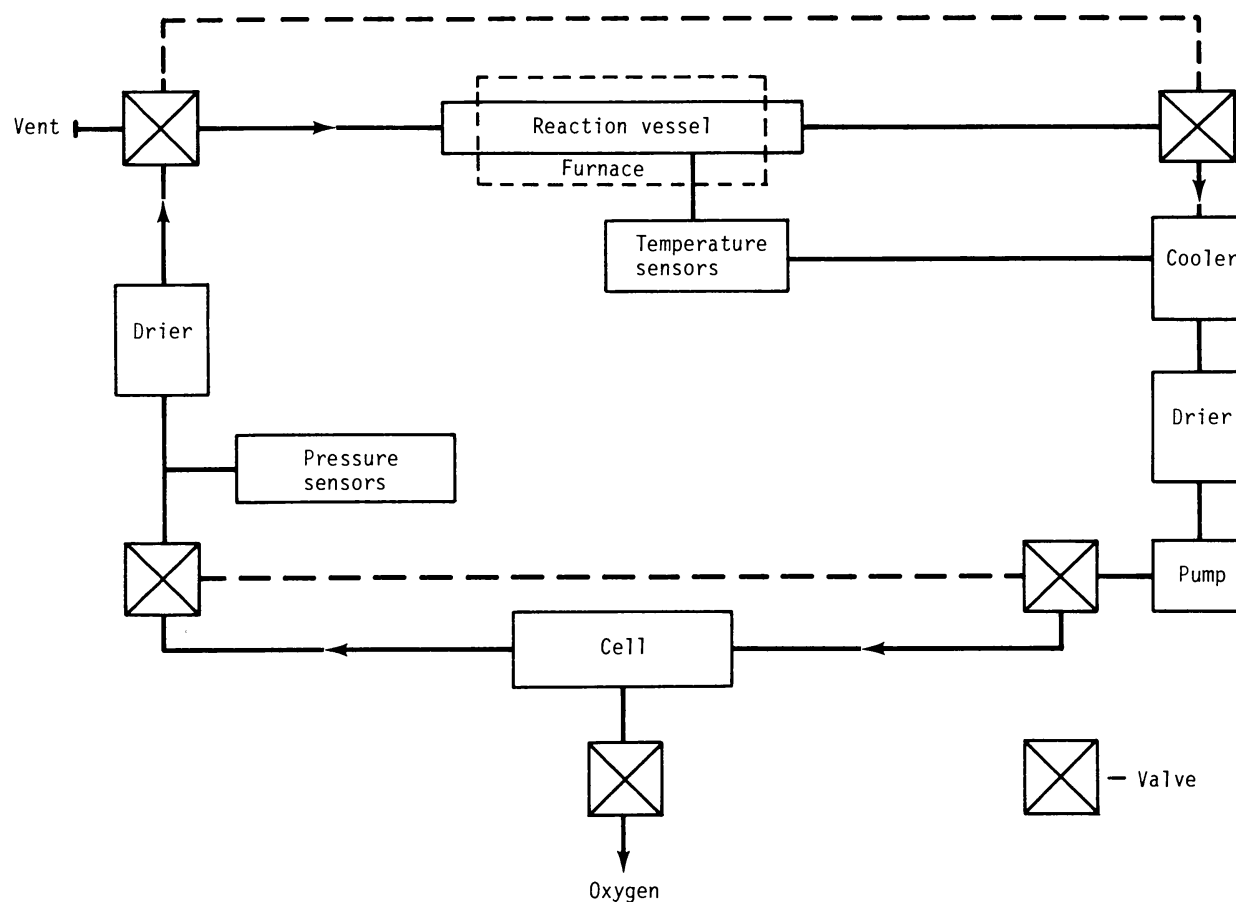


Figure 1. Schematic of Oxygen Production Test System. Arrows show direction of gas flow. Dashed lines are bypass used during purge of system. Power, control, and data acquisition subsystems are not shown.



## EXPERIMENTAL APPARATUS

The apparatus (see Fig. 1) used for the test was constructed from commercial components with the exception of the reactor vessel that was designed and fabricated in-house and driers that were constructed from nylon rod. Stainless Swagelock or Cajon-type fittings were used throughout the system (which was constructed using stainless tubing). The reactor vessel was fabricated from 304 stainless steel and used a copper gasket seal; it was designed to ASME Boiler and Pressure Vessel Code specifications (Section III, division 2) for 1500°F and 100 psi. The vessel had an outside diameter of 1-1/2", a bore of 3/4", and was 11" long. The furnace was a commercial nichrome-wound tube furnace controlled by a thermocouple-driven proportional controller. The cooler was a spiral of copper tubing soldered to a bank of four thermoelectric coolers; an instrument cooling fan provided air circulation for removing heat from thermoelectric devices. The pump was a light duty circulating pump that provided about 2 psi pressure head. Finally, the cell was a General Electric hydrogen generator with the electrolysis membrane reversed so that hydrogen was released on the "dry" side of the system. The masses of the subsystems and the total system volume is given in Table 2.

The temperature, pressure, and the power consumption of the various active components were measured during testing. The small quantities of water produced during the test could not be recycled and measured simultaneously without recourse to extremely long experiments. Thus, yields were measured by absorbing the water produced and comparing the yields from experiments with and without a cooler.

## TESTING AND DATA

Results from the following tests are the basis of this paper:

1. Hydrogen flow, cooler on,  $\text{Al}_2\text{O}_3$  granules in the reactor
2. Hydrogen flow, cooler off, ilmenite in the reactor
3. Hydrogen flow, cooler on, ilmenite in the reactor

Table 2. Component Masses

Components	Mass (Kg)
Pump	3.5
Furnace	2.7
Vessel (empty)	0.8
Electrolysis cell	2.0
Power supply for cell	3.0
Condenser	1.1
Power supply for condenser	3.9
Tubing, miscellaneous	1.5
Total volume of closed loop (including vessel and filters)	
202 cm <sup>3</sup> .	

Table 3. Chemical and Mineralogical Analyses

	Wt %
SiO <sub>2</sub>	2.72
TiO <sub>2</sub>	38.75
Fe <sub>2</sub> O <sub>3</sub>	54.21
MnO	0.45
MgO	2.02
CaO	0.39
SO <sub>3</sub>	0.90

Mineralogical Analysis: X-ray analysis indicates that the sample is a mixture of ilmenite with calcic plagioclase, olivine, and hornblende. SEM analysis of the ilmenite indicates a composition of  $\text{Ilm}_{97}\text{Hm}_3$ . It also revealed the presence of a minor sulfide with the composition FeS.

Nominal test conditions were:  $P_{\text{H}_2} = 10$  psi; reactor temperature = 705°C; condenser temperature = 3°C. The system, set for bypass (see Fig. 1), was thoroughly dried and tested for leaks using helium flow between tests. The vessel contained approximately 90 g of ilmenite (black, massive variety from Quebec; see Table 3 for chemical and mineralogical analysis) that had been crushed, sieved to less than 150 microns, and maintained in a vacuum oven at 80°C until used. The ilmenite was not changed between Test 2 and 3. Water yield was measured by weighing the drier (loaded with "drierite") before and after the tests, each of which was nominally one hour in duration. The data from the three tests are shown in Table 4. Voltages and current measurements indicate that about 2400 watts were required to maintain the system under operational conditions (see Table 5 for breakdown by subsystem).

Table 4. Test Results

Weight	Gain	Duration
Blank, cold trap	3 mg	49 min.
Ilmenite, no cold trap	15 mg	63 min.
Ilmenite, cold trap	65 mg	76 min.

Reactor temperature =  $703 \pm 5^\circ\text{C}$ .

Cold trap =  $3 \pm 2^\circ\text{C}$ .

Room temperatures =  $22 \pm 0.5^\circ\text{C}$ .

Note:  $\pm$  are observed fluctuations; values are average of measured values.

Table 5. Power Consumption

Source	Watts
Furnace	1475
Electrolysis cell	677
Cooler	89
Pump	77
Sensor, etc.	86

Note: Values are averages of measured values.

## ANALYSIS

Table 6 presents the yield of water (mg/min) and compares Test 2 (no cold trap) with Test 3 (cold trap). It is assumed that the yields are linear with time, and that the minor variations in parameters from experiment to experiment and within a given experiment as a function of time do not significantly affect the results. The data indicate that yields and the specific yields (yield/unit mass and yield/unit power) are all higher by a factor of 3–4 for the test with cooling versus the equivalent test without cooling. Thus, enhancements noted by Williams and Mullins (1983) for batch operation are evident in continuous mode, although they are not as large as the factor of 20 suggested by theory.

## SUMMARY

The enhancement of water production from the hydrogen reduction of ilmenite by the use of cold traps has been demonstrated for a continuous flow process. However, the improvements were not as dramatic as those predicted from theory or those obtained in batch operation. The result is not particularly surprising, since the calculated enhancements will apply only to a system of overall equilibrium—that is, only static systems, like batch processing, will show the full benefit of cold-trapping. It is significant that some enhancement has been noted for the continuous flow process, since this suggests that an optimal continuous flow process may exist in which the benefits of enhanced yield can be combined with those of continuous processing. Studies to define this optimum would be useful.

Table 6. System Performance

Test No.	Yield (mg/min)	Power (W)	mg/KW-Hr	Mass (Kg)	mg/Kg-min
2	0.18	2315	6.17	13.5	0.013
3	0.80	2404	21.34	18.5	0.043

Notes: Yield is net from Table 2 minus baseline (0.06 mg/min). Mass is from Table 1; condenser and its power supply mass have been removed from mass budget for Test 2.

The current data are limited by the number of tests performed and duration of those tests. However, further testing would require significant redesign of the experiment in two ways:

1. The relative size of the electrolysis cell and the reaction vessel need to be better matched. The cell used in these experiments is at least a factor of 10 too large; consequently, an indirect (water absorption) technique had to be used to measure production. A much smaller cell or a much larger vessel would have permitted direct observation of water levels during the experiment.
2. The terrestrial ilmenite used in these studies contains minor sulfide. The  $H_2S$  produced by the reaction of the sulfides with hydrogen attacked the electrolysis cell membrane; it was this that forced termination of testing. Lunar ilmenite is also associated with sulfides. Thus, any process will have to deal with these contaminants, using physical or chemical techniques to purify starting materials or efficient gases. Technologies (see, for example, King, 1974) exist that can handle such problems; however, these add complexities and costs to the system that have not been fully appreciated.

We believe that the test program described in this paper was successful in demonstrating the principles of operation, quantifying yields, and isolating design problems for future researchers. However, it must be emphasized that we were mainly dealing with feasibility and preliminary concept testing; thus, we could be sloppy to some degree—for example, the presence of ferric iron in our ilmenite is not critical. The next generation of tests will require more sophistication and a more complete simulant to obtain quantitatively relevant process data. When better simulants are available, further testing should proceed to explore the behavior over longer times and to optimize production. Hopefully, our experience is a useful introduction to such research.

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# **A CARBOTHERMAL SCHEME FOR LUNAR OXYGEN PRODUCTION**

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Carbothermal reduction may be the process of choice for winning oxygen from lunar materials. A wealth of terrestrial engineering experience can be applied to development of a lunar plant. Coproducts, such as slag and steel, can be utilized in lunar operations and may be useful lunar exports. Carbon is a more efficient reductant on a per mass basis than is hydrogen in the competing ilmenite reduction process. An accurate plant design awaits further research to address uncertainties in process kinetics, behavior, catalysts, and reactor materials.

## **INTRODUCTION**

The most useful material to produce on the Moon is oxygen for propellant (Cole and Segal, 1964). During the Apollo flights, about 75% of the Saturn V's effort placed propellant in lunar orbit and on the lunar surface to enable return to Earth. A lunar source of oxygen could double or triple the net lunar base mass (and capability) for a given Earth to LEO mass throughput. It also can provide inexpensive gas for a number of uses in lunar exploration. Lunar oxygen may even be exported to LEO at costs arguably below Earth to LEO launch costs (Salkeld, 1966; Davis, 1983; Andrews and Snow, 1981; Cutler, 1984; Cutler and Hughes, 1985; Simon, 1985).

Transportation requirements and transportation economics for the utilization of lunar resources in Earth orbit are reasonably well understood (although market models are somewhat ill-defined). However, the actual production of basic commodities from available lunar minerals is poorly understood (Duke and Arnold, 1977; McKay and Nozette, 1985). The lunar environment is sufficiently different from the terrestrial environment that plant design and process chemistry will differ substantially from that used on Earth. Currently no accurate plant designs exist that can be used in systems studies or mission definition.

A candidate approach for winning oxygen from lunar minerals is the carbothermal process (Fig. 1), which yields steel as a necessary by-product. The process combines chemistry from steel-making and from coal synthesis gas reforming with electrolysis or thermolysis of water. Research was carried out on the carbothermal reduction process in the early 1960s and was reported in the technical literature and NASA technical reports (Rosenberg *et al.*, 1963a,b; Rosenberg *et al.*, 1964a,b,c,d,e; Rosenberg *et al.*, 1965a,b,c,d,e,f).

Every step in this process has been used extensively in engineering practice. The terrestrial data provide an excellent framework for research on and possible development of this process for the extraterrestrial environment.

The near-term benefits of lunar oxygen are recognized. Lunar steel may also be a near-term useful product. It could be utilized in many lunar base components such as roadbeds, landing pads, instrument foundations, walkways, stairways, pressure vessels, pipes, drill rigs, and cables. A source of lunar steel combined with lunar base propellant production could enable various large scale space projects. If we continue to transport all needed materials from Earth, space activities in the 21st Century will be as they are now—limited and expensive. We must learn to use space resources if we are to change our presence in space from tentative peeks to vigorous exploration and exploitation. The carbothermal processing plant is a good example of a technology that takes maximum advantage of terrestrially derived engineering knowledge to start using extraterrestrial resources.

## PROCESS CONSIDERATIONS

The carbothermal process for producing lunar oxygen (Fig. 1) starts with mining regolith and separating out a desirable mineral fraction, probably ilmenite. About 100,000 tons of lunar regolith must be mined per year to produce 1000 tons of oxygen, based on a 10% usable ilmenite content. This corresponds to digging a pit 100-m × 100-m

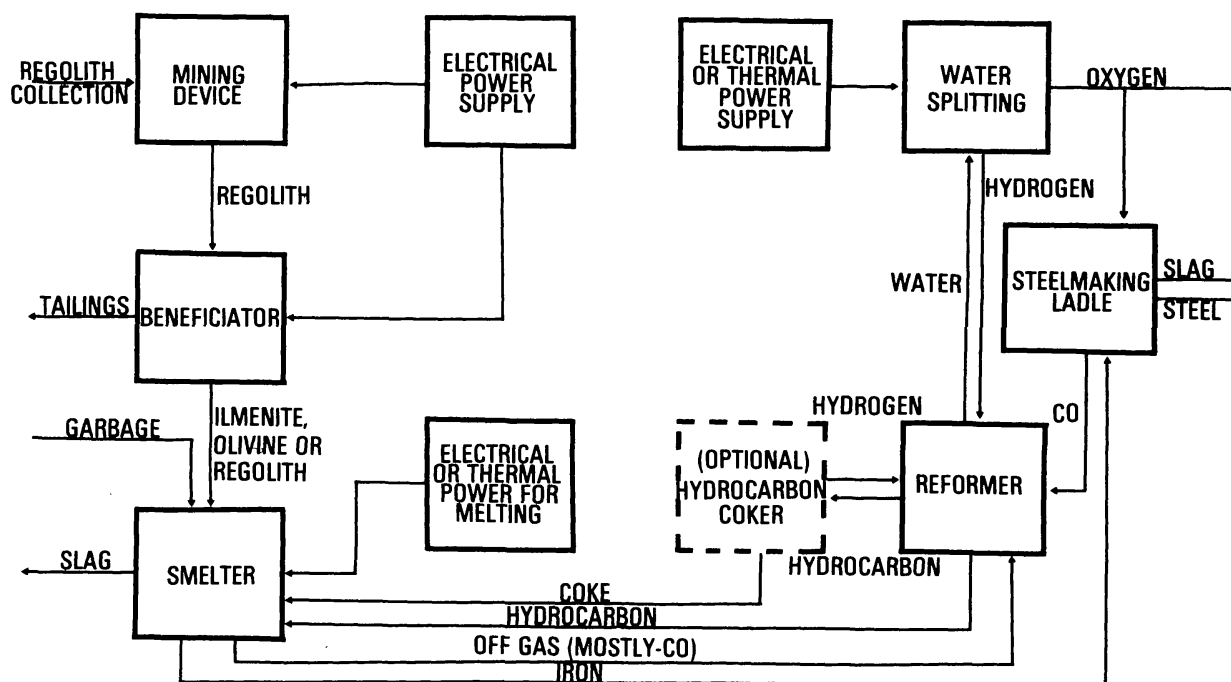
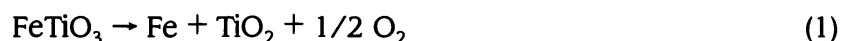


Figure 1. Flow diagram of the carbothermal oxygen production plant. Regolith is mined; steel and oxygen are produced; and tailings, iron-making slag, and steel-making slag are discarded.

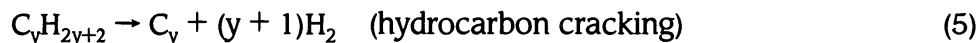
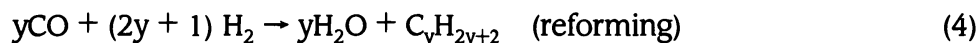
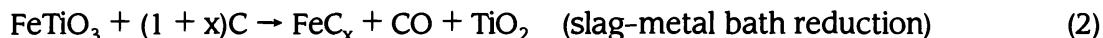
× 5-m deep over the year, about the amount of material excavated in building 1.5 miles of interstate highway. A beneficiator removes oversize material and separates the remainder into tailings and feed material, about 90% ilmenite combined with 10% flux. The tailings are disposed of down a conveniently located slope, and the feed materials charged to the smelter.

In the proposed lunar system, the feed contains ilmenite, anorthite as a fluxing agent, and any carbon-bearing solids garnered from garbage or the off-gas stream. Phase diagrams of the system,  $\text{TiO}_2 - \text{SiO}_2 - \text{Al}_2\text{O}_3 - \text{CaO}$  (Nurnberg, 1981), show that a 10% addition of anorthite is sufficient to lower the slag melting point below that of the iron product.

The overall process chemistry in the plant is:



This is composed of the following steps (not showing partial reduction of  $\text{TiO}_2$ ):



In the smelter, feed material is mixed with a carbonaceous reductant and heated until melting, reduction, and slag/metal separation take place. The reductant should be as hydrogen poor as possible to keep the volume of smelter off-gas down and to reduce heat loss in this stream. The reductant is recycled carbon, reformed from monoxide to waxes by Fischer-Tropsch synthesis and is perhaps cracked (4,5). Unfortunately, these waxes do not coke well, and research and development may be required to improve the yield of crackable products. The light ends can be reformed either by partial reoxidation or arc cracking to give materials more suitable for coking, at the cost of substantially increased power consumption.

Carbon is the refractory of choice for this reactor due to its high strength to weight ratio, its resistance to attack by the (titanium carbide saturated) iron product, and its availability from electrode wear, garbage, and the recycled off-gas. Carbon is used in terrestrial ilmenite smelters and, with the slag skull, is found to be semi-permanent in service (Noda, 1965). Some operating data are available (Knoerr, 1952; Noda, 1965; Grau and Poggi, 1978) from plants that use a thermal gradient stabilized slag skull to protect the refractory lining from chemical attack. Process power consumption is about 3.5 MWh/T Fe (megawatt-hours per ton of iron), and arc heating electrode consumption is between



5 and 10 kg/T Fe, which translates to converting about 0.75% of the carbon flow in the total process into electrodes. Arc heating uses low voltage DC power like that generated in photovoltaic or nuclear thermionic processes.

Substantial research has been done on processes for iron production by introducing hydrocarbon, coal, or coke into a slag-metal bath (Eketorp *et al.*, 1981). Because reduction in the bath (2) is an endothermic process, it has not proven possible to get rapid reaction on the scale of typical terrestrial iron reduction furnaces (10,000 tons of hot metal produced per day). However, calculations indicate that heat transfer in the bath is more than adequate for the small scale of operation relevant to lunar iron reduction plants.

One advantage of smelting ilmenite is that under process conditions the titania will be partially reduced. This increases oxygen recovery per unit throughput and decreases the amount of iron that must be decarburized per unit oxygen produced. However, feed minerals for lunar iron production have low iron content compared to terrestrial ores, causing a high ratio of slag to iron and increasing the heat demands and reactor size per unit oxygen (or iron) production.

The pig iron product will contain carbon, and decarburization (3) is required to make a usable product and to obtain high carbon recycle efficiency. Decarburization could employ steam, oxygen, or wet hydrogen. Low lunar gravity is helpful to decarburization by allowing longer gas/liquid contact times. Steam decarburization is endothermic and thus would not lead to hot spots in the bath or the severe refractory wear associated with oxygen decarburization. Alternatively, oxygen decarburization is exothermic, so the vessel would need no heat source. The overall oxygen/steam ratio could be adjusted to bring the reactor into thermal balance. Although power availability and materials arguments imply that consumable electrode arc heating would be the most practical approach to heat introduction, induction heating, which also removes the need for hydrocarbon cracking and electrode production, is possible. Modern induction power converters have masses ranging from 2 metric tons at 1MW to 3.5 metric tons at 2MW. Conversion efficiency is typically 96%, adding 40–80 kW to the heat load that must be rejected at low temperatures.

Counter-current heat exchange between pelletized feed and reactor off-gas in a moving bed could decrease the electrical energy requirement. It would also cool the off-gas to suitable temperature for subsequent processing and would condense volatile impurities.

Both the decarburization and reduction off-gases will contain sulfur that could poison the Fischer-Tropsch catalysts. The best solution is to use resistant catalysts, or else to pass the off-gases through a water spray made basic by alkali oxide fumes. (This water spray is a small added inventory of water on its way to the electrolyzer.)

Water is a by-product of Fischer-Tropsch synthesis; its electrolysis (6) leads to the product oxygen, which is liquified and stored. Electrolysis has been extensively studied in a space context. Literature also exists on using thermal cycles for production of hydrogen and oxygen from water, often using solar heat. Thermal water-splitting, if available, would significantly reduce electrical power demands. Although processes making direct use of solar radiation are known (Raissi and Antal, 1985), further research and design studies are needed.

## SYSTEM CONSIDERATIONS

A variety of considerations apply to any system involving the production of oxygen on the Moon and its delivery to Earth orbit. Tradeoffs must be made involving solar versus nuclear power supply and continuous versus batch processing. Systems design and economic criteria will be quite different if a lunar oxygen producing plant is installed before a lunar base or as part of a lunar base program.

A coexisting lunar base would have several beneficial effects on lunar factory operations and economics, since routine and non-routine repairs could be attended to quickly and would be a marginal cost (the marginal cost of extra person-hours) rather than a direct cost with the addition of transport charges. Propellant oxygen for transportation from the Moon is an economical product even under circumstances where lunar oxygen export to Earth orbit may prove uneconomical. Inexpensive plant by-products, such as cast slag bricks or formed steel parts, may have some utility and economic value at the lunar base.

Without a coexisting base, reliability would become very important and would have to be ensured even at a high cost in mass or initial factory cost. One simple means of enhancing product supply reliability is to install over-capacity, so that unexpected downtime does not deplete product stores. Another approach is to have redundant plant elements so that unscheduled maintenance does not stop all production. The costs and benefits of over-capacity are obvious. The change in capacity required for reasonable reliability (10–20%) is not enough to affect the plant systems design substantially. Redundancy may be a different story because the total capacity is shared by multiple small plants. Since plant masses (and capital costs) scale according to the  $2/3$  power of capacity, the ratios of costs for no redundancy, dual redundancy, and triple redundancy are 1:1.26:1.44.

For a carbothermal oxygen plant, the reduction in capacity of each process reactor with increased redundancy means that the smelting reactor will become more difficult to operate in a continuous mode. The costs of batch and continuous processes scale differently, with large scale favoring continuous processing. Continuous operation is very desirable on the Moon due to the ease of automation and automatic control, simplified design, lower thermal cycling, and facile use of gravity feed when compared to batch processing. If the capacity reduction needed to make redundant oxygen factories forced a transition from continuous to batch smelting, the capital cost penalties would increase substantially. Electrolysis, reforming, and cracking units are operated economically on Earth in continuous mode on a scale similar to that necessary for a lunar factory capable of producing 1000 tons of oxygen per year. Despite possible increased costs, redundancy is very effective in reducing the likelihood of unexpected supply interruptions if a plant must be operated at an unmanned lunar facility. Single versus redundant plants must be traded off in any systems study of lunar oxygen production at an unmanned facility.

Continuous process ilmenite smelters are operated on Earth at 10–20 times the capacity needed in a 1000 ton per year of oxygen lunar factory, but the thermal balance in these

Table 1. Energy and Power Requirements per Ton of Product Oxygen

Process Step	Energy (GJ)
Reduce 3.68 tons of iron (75% efficiency)	12.1
Heat to melt 3.68 tons of iron (500 kWh/ ton)	6.6
Heat content of 4.25 tons of slag (470 kWh/ ton)	7.2
Heat content of off gas (1350 oC effective heating)	8.0
Energy to electrolyze water (60% efficiency)	28.9
Energy to liquify oxygen	5.4
Total energy consumption, carbothermal process	68.2
Total power requirement for 1000 tons of O <sub>2</sub> per year (100% duty cycle)	2.16 MW
Nuclear power plant capacity (90% duty cycle)	2.40 MW
Solar power plant capacity (40% duty cycle)	5.40 MW

Methane reductant, no heat regeneration.

Data scaled from that presented by Rosenberg *et al.* (1965f) for a 12,000 lb/mon oxygen production facility without heat pumping. Power was assumed to scale linearly.

smelting units is delicate. The possibility of scaling a continuous smelting reactor down to the capacity needed for carbothermal production of lunar oxygen must be the subject of further study.

In contrast, all terrestrial steel-making is done in batch processes. Capacities up to several hundred tons of steel per hour are common. It seems unlikely that lunar steel-making can be done in a continuous mode.

Manufacturing plant power demand will be several megawatts (Table 1), well in excess of power demand for other functions of a lunar base. Nuclear energy is likely to be used to power a manned surface facility, due to the difficulty of storing solar derived energy for use during the lunar night. Continuous energy supply may not be a requirement for a lunar factory. Since factory power demands are much higher than and have a different time phasing and quality from other power demands, a total systems design will be required to select the optimum power system for the complex. In fact, the factory and base may have separate power systems.

There are two competing factors in the tradeoff between solar and nuclear energy for the lunar factory power supply: (a) the relative power to mass ratios of solar and nuclear power systems, and (b) the overall plant masses required to produce a given amount of product when the power supply and plant duty cycles are taken into account. Plant mass breakdowns for solar and nuclear powered plants are given in Table 2. The simple assumptions used in this table give solar and nuclear powered plant masses that are essentially equal. Accurate projections of solar and nuclear power system power to mass ratios as well as accurate scaling laws derived from careful design studies will be needed before any decision can be made about the selection of power sources.

The delivery of a lunar oxygen factory to the Moon's surface and the storage and transportation of lunar oxygen are separable from the lunar oxygen manufacturing plant itself in a systems sense. Transportation system characteristics are more likely to be

Table 2. Processing Plant Mass per Unit Output of Product

Product	100% Duty Cycle		90% Duty Cycle		40% Duty Cycle	
	Oxygen	Iron	Oxygen	Iron	Oxygen	Iron
Processing plant	30.4	8.69	32.6	9.32	56.0	16.0
Mining and beneficiation	10.8	3.09	12.0	3.43	27.0	7.7
Oxygen plant mass			44.6		83.0	
Power plant (Table 1):						
Nuclear (40 W/kg)			60			
Solar (190 W/kg)					28.5	
Total mass			104.6		111.5	

In tons per 1000 tons per year.

Data scaled from that presented by Rosenberg *et al.* (1965f) for a 12,000 lb/month oxygen production facility without heat pumping. A decarburization reactor (taken as equal in mass to the reduction reactor) was added. Decarburization is assumed to consume 5% of the ultimate O<sub>2</sub> production. Plant mass is assumed to scale as the 2/3 power of capacity.

determined by space station and lunar base needs than by lunar oxygen factory needs. Nevertheless, there exists a design interaction between maximum size and mass of factory components and the payload capacity of the transportation system.

## DISCUSSION

The carbothermal reduction process has been discussed in an appropriate thermodynamic context, and plant mass and power estimates have been made (Rosenberg *et al.*, 1965f). Mass estimates (Table 2) are 32.6 tons of plant and power requirements equal to 60 tons of 1990 (projected) nuclear power supply and mining and beneficiation equipment of about 12 tons per 1000 tons of oxygen production per year. Mining equipment mass is taken from Gertsch (1983), and beneficiation is assumed to be similar to that reported by Agosto (1985). Assuming a mass ratio of iron to oxygen of 3.5:1 (as for stoichiometric oxide), these correspond to 9 tons of plant supply, 17 tons of power supply, and 3 tons of mineral handling equipment per ton of steel produced per day in an ilmenite smelting process.

The carbothermal process for lunar oxygen production and a process based on the hydrogen reduction of ilmenite bear the same relationship to each other as direct reduction and smelting do in terrestrial engineering. It is thus reasonable to expect them to be competitive technologies as they are on Earth.

The hydrogen reduction process reacts the mined ilmenite with hot hydrogen to produce water, iron, and rutile. The water is electrolyzed to produce oxygen and regenerate hydrogen. Thermodynamic equilibrium limits the per pass conversion of hydrogen to water to about 5%. Subsolidus hydrogen reduction of ilmenite produces an intimate mixture of iron and rutile and would require a subsequent thermal processing step to separate the iron.

There are several factors that seem to favor the carbothermal reduction based process. Makeup carbon is available from a variety of sources, such as garbage and scrap. Hydrocarbons and carbon are much easier to handle than hydrogen. This is particularly significant when some reagent inventory must be kept on hand for makeup between supply deliveries from Earth. As process temperature increases, significantly more oxygen can be recovered from a given weight of ilmenite. Reduction carried out under slagging conditions will only require 1/2 to 2/3 as much ilmenite per unit oxygen recovery as would subsolidus reduction at 700°C–900°C.

The major problem with hydrogen reduction of ilmenite appears to be the unfavorable equilibrium constant for the conversion of hydrogen to water. This ranges from 0.031 at 600°C (Williams, 1983) to 0.117 at 1300°C (Shomate *et al.*, 1946). If the hot, wet hydrogen is cooled down to condense out the water and then reheated, an excessive heat demand is imposed on the system. Developing a high temperature electrolysis cell using a ceramic oxide ion conductor to dewater the hydrogen without cooling it adds development costs and risks. In contrast, carbon reacts completely in a single pass. It also extracts more oxygen from ilmenite in a single pass than does hydrogen on a mass basis. (A mass unit of carbon extracts 1.33 to 1.45 mass units of oxygen per pass, while a mass of hydrogen extracts 0.25 to 0.94 mass units of oxygen per pass, depending on the equilibrium conversion limit.)

The following processes that have been proposed to extract oxygen from lunar material cannot be compared directly with the carbothermal process but are considered to have significant technical weakness:

1. Electrolysis of molten lunar minerals has been studied as an oxygen producing process. However, no anode material tried to date has clearly demonstrated adequate corrosion resistance (Haskin and Lindstrom, unpublished data, 1984), leaving some possibility that the process is impractical.

2. Vapor phase pyrolysis of lunar minerals involves vaporizing lunar soil or selected mineral separates and then rapidly quenching the vapor. The hot gas contains some metal atoms and oxygen molecules. Literature data (Borgianni *et al.*, 1969) make it seem unlikely that quenching of the hot gas can be made rapid enough to prevent loss of oxygen through reoxidation.

3. The hydrofluoric acid leach process as described by Waldron (1985) involves dissolving bulk lunar soil in a fluoride based acid, separating the resulting mixed salt solution into pure metal fluorides, reducing these metal fluorides with sodium and potassium, hydrolyzing the resulting alkali fluorides to regenerate hydrofluoric acid, and producing oxygen by electrolysis of molten alkali hydroxides. Design-based mass estimates are 77 tons of plant per ton of soil input per day, or 586 tons of plant per thousand tons of oxygen per year (assuming 40% oxygen by mass of lunar soil and a 90% duty cycle), much higher than for competing processes.

## RESEARCH NEEDS

Conceivably, fatal flaws could exist in the carbothermal process in any of three areas where research is badly needed: (a) kinetics and phenomenology of (simulated) lunar

ilmenite reduction, (b) carbide solubility in typical slags, and (c) iron decarburization. Experimental data on these key issues would narrow design uncertainties in the two least well understood parts of the system, the smelter and the steel-making reactor.

Once it has been verified that the basic process chemistry for carbothermal production of lunar oxygen is sound, the following secondary research projects would define system performance and design. A Fischer-Tropsch catalyst is needed that gives readily crackable product with a low H to C ratio and which is also economical in the lunar context. Appropriate hydrocarbon coking and electrode production techniques need development. Thermal methods of producing hydrogen and oxygen from water should be explored and traded off against electrolysis in system design studies. Various methods of heat rejection and thermal control should be studied and compared to each other at the system level. All of the above studies would have to be performed to determine whether plant development is warranted.

The choice between a nuclear or a solar powered system depends on the relative power-to-mass ratios and on the plant mass scaling law. The tradeoff seems close now and cannot be resolved without careful systems studies shortly before plant emplacement.

Separating water into hydrogen and oxygen by thermolysis is an interesting possibility for a carbothermal process plant and for a variety of other types of plants. Water thermolysis should be examined in a space context independently of other projects.

A lunar oxygen factory in the context of a lunar base program can be the source of inexpensive by-products (slag and steel) that would be very useful to the base. The properties of slag or steel manufactures cannot be predicted solely on the basis of compositional data. If these by-product materials were to be useful in lunar base activities, their properties would have to be determined experimentally using simulants of appropriate composition.

If development seems to be a possibility, the interface between the oxygen producing factory (after the oxygen liquifier) and the storage and transportation system will have to be studied. If the oxygen storage facilities on the lunar surface are considered part of the transportation system, parameters such as mission frequency have practically no effect on the oxygen producing plant, and few plant parameters have any effect on the transportation system.

## **CONCLUSIONS**

Carbon is an economical reductant in the lunar context because it removes more than 1.25 times its mass of oxygen per pass. Hydrogen, though lighter per mole, only removes 0.4–0.9 times its own mass per pass due to thermodynamic limitations. Makeup carbon might be readily available from spent factory delivery stages, scrap, or lunar base garbage.

Thus the carbothermal route to lunar oxygen is economically and technically attractive. There is low technical and performance risk in plant design and development due to extensive Earth-based experience with each element of the process. The basic research needed to demonstrate the feasibility of this concept is straightforward and well defined. It consists of quantifying carbon loss in slag and steel and studying process behavior

in the reduction reactor. Further definition should be pursued, both in the laboratory and through selected system studies.

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# LUNAR REGOLITH FINES: A SOURCE OF HYDROGEN

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The theoretical evaluation of the lunar regolith fines as a primary source of hydrogen reveals that a minimum order of magnitude increase in hydrogen content is possible in beneficiated fines because both particle size and particle shape play a significant role in the relationship of volume percent of surface coating to grain size. The lunar regolith fines meet the basic requirement for beneficiation because a major portion (minimum two-thirds) of the hydrogen occurs in the less than 20- $\mu$ m-size fraction, a relatively small part of the fines. Beneficiation should be accomplished by a combination of vibratory screening followed by cyclone and/or possibly electrostatic separation. Early exploitation of the lunar regolith fines for hydrogen probably will be limited to hydrogen obtained as a by-product or co-product from the mining and processing for other elements or materials because a minimum of about 13,600 tons to about 19,600 tons of 100 ppm hydrogen-bearing lunar regolith fines will have to be processed with about 3,100 tons to about 4,500 tons, respectively, of concentrate heated to supply 1 ton of hydrogen, yielding a recovery of about 74% to about 51%, respectively, of the hydrogen.

## INTRODUCTION

The interest in establishing a permanent manned presence on the Moon makes it imperative to determine if indigenous lunar material can be mined and extracted economically to provide materials needed for a lunar base operation (McKay and Williams, 1979). Importing materials to the lunar surface from Earth initially will cost approximately \$25,000 per pound in 1984 dollars—over five times the value of gold on Earth! This paper (a) explores the possibility of the lunar regolith fines as a primary source of hydrogen—a critical element from the viewpoint of consumables (e.g., propellant, water, and hydrogen-bearing reagents) and (b) compares the known lunar data with the developed hypothesis.

The known concentration range of hydrogen in lunar regolith fines is shown in Table 1. Such values are often used as evidence that the Moon is devoid of water even though 100 ppm of hydrogen is equivalent to 0.09 wt % water. It would, however, take 10,000 tons of 100 ppm hydrogen-containing lunar regolith fines to produce 1 ton of hydrogen at 100% recovery. Therefore, if lunar regolith fines are to become an economical source for hydrogen, they must be beneficiated because at least 99.98% of this type of lunar material is “waste” (Table 1) and would consume energy during the extraction of hydrogen. Beneficiation of lunar regolith fines can only occur if a relatively small portion of the material in the fines contains a significant amount of the hydrogen and also if that material can be separated, which implies it must have unique physical or chemical properties. In addition, the process must be economical, that is, not be labor intensive or technically complex.

Table 1. Range of Hydrogen Content of Bulk Lunar Regolith Materials in ppm from Apollo Missions

Apollo Mission	Hydrogen (ppm)
11	38– 60
12	26– 80
14	61–106
15	15–120
16	10– 79
17	17–211

Data from Taylor (1975); Bustin *et al.* (1984).

## PREVIOUS MODEL STUDIES

Implanted solar wind particles, of which hydrogen is the most abundant component (Cameron, 1973), are known to penetrate to depths greater than 1000 Å, but most of the particles are trapped within 200 Å of the surface (Leich *et al.*, 1973, 1974; DesMarais *et al.*, 1974). DesMarais *et al.* (1974) recognized both surface- and volume-correlated components of hydrogen in lunar fines by means of analyses of the distribution of hydrogen with respect to particle size assuming spherical shapes. Bogard (1977) developed the idea that solar wind implanted gases become increasingly volume-correlated by incorporation into agglutinates as a soil matures. Frick *et al.* (1975) developed the concept of spherical shells containing an equilibrated gas concentration to allow for a considerable penetration depth of an incident noble gas particle. However, the abstract by Carter (1984) is the first attempt to quantitatively evaluate the relationship of volume of surface coating to particle size or shape.

## THEORETICAL CONSIDERATIONS

### General

Consider a perfectly spherical soil grain of diameter  $D$  with a gas-enriched outer layer of thickness  $t$ . The volume of the enriched layer is related to the total volume of the grain by the formula:  $V_{\text{layer}} = V_{\text{grain}} [6(t/D) - 12(t/D)^2 + 8(t/D)^3]$ . If the thickness of the layer is small compared to the diameter of the grain, then the fraction of the volume that is enriched can be estimated adequately using only the first term of the expansion. For example, a 200-Å coating on a 20-μm grain would represent an enrichment of only 0.6% of the total grain volume. Since large grains would require very thick coatings to provide any significant enriched volume, all subsequent discussions will be confined generally to data for particles 20 μm or less in diameter.

### Surface Coating Volume Relationships

The importance of both grain diameter and thickness of coating to volume percent of surface coating on spheres is shown in Fig. 1. A significant amount of coating volume

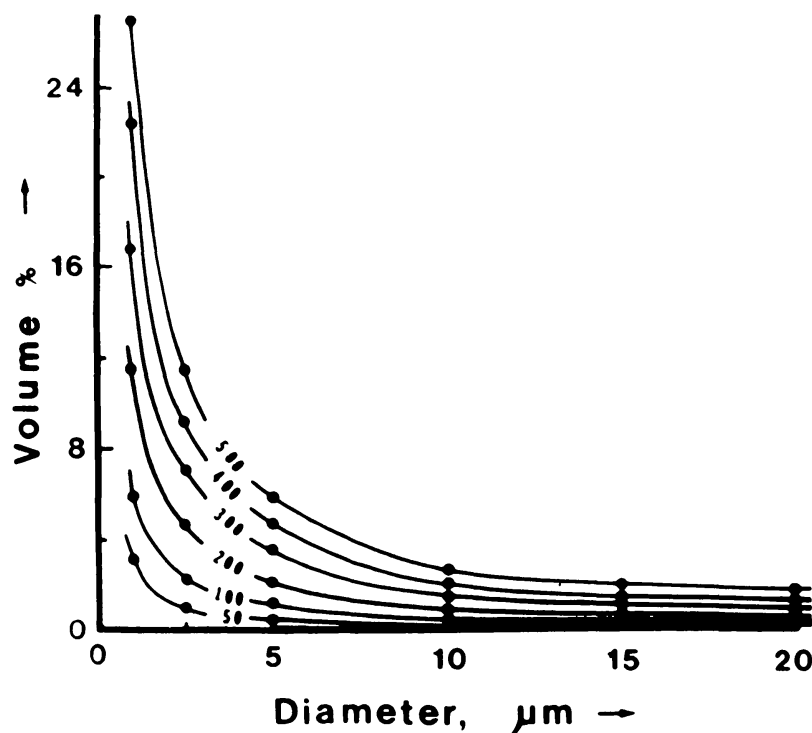


Figure 1. Relationship of volume percent of surface coating versus diameter in microns for coating thicknesses from 50-500 Å on spheres.

(greater than 1%) occurs (a) on particles 20  $\mu\text{m}$  or less in diameter for all surface coating thicknesses greater than about 334 Å and (b) on particles with diameters less than about 3  $\mu\text{m}$  for all surface coating thicknesses of at least 50 Å.

The volume percent coating on spheres versus surface coating thickness up to 200 Å for various particle diameters is shown in Fig. 2. The intersection of the dashed horizontal line with each labeled particle diameter line reveals the minimum coating thickness

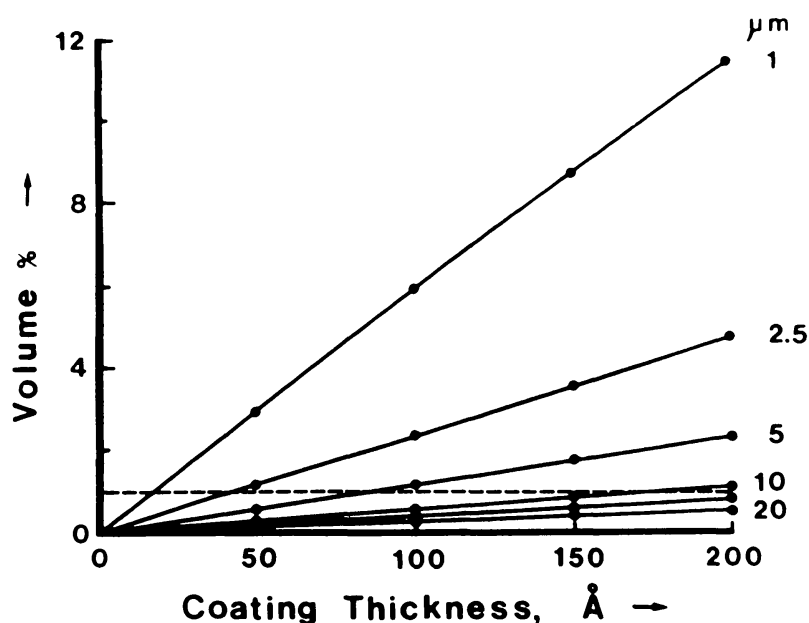


Figure 2. Relationship of volume percent of surface coating on spheres versus coating thickness in angstroms for particle diameters of 1-20  $\mu\text{m}$ . Area above dashed line represents volume percent of surface coatings greater than 1%.

Table 2. Relationship of Percent of Surface Coating Volume Versus Maximum Particle Length in  $\mu\text{m}$  for 200-Å-thick Surface Coating on Platic and Prismatic Shapes, Where the Maximum Length is Twice the Shortest Length

Maximum Length, $\mu\text{m}$	% Volume	
	Plate*	Prism**
1.0	15.2128	18.7456
2.5	6.2728	7.7968
5.0	3.1681	3.9490
10.0	1.5920	1.9872
15.0	1.0631	1.3277
20.0	0.7980	0.9968

\*% Volume =  $100\{[(A)^2 \times 0.5A] - [(A-0.04)^2 \times (0.5A-0.04)]\} / [(A)^2 \times 0.5A]$ ; where A = maximum particle length.

\*\*% Volume =  $100\{[(A) \times (0.5A)^2] - [(A-0.04) \times (0.5A-0.04)^2]\} / [(A) \times (0.5A)^2]$ ; where A = maximum particle length.

necessary for a coating of 1 vol %. For example, for a 2.5- $\mu\text{m}$ -size grain a minimum thickness of surface coating to give 1 vol % is 42 Å. Figure 2 also can be used to give the maximum grain size in micrometers for a given thickness and a given volume percent of surface coating. For example, for an average 200-Å-thick coating on spheres, the volume of coating is greater than 1% for all grain sizes less than 12  $\mu\text{m}$  in diameter.

### Particle Shape Relationships

The theoretical effect of grain shape on volume of coating to grain diameter has not previously been systematically examined (Carter, 1984). The importance of grain shape on the volume relationships is demonstrated by Tables 2 and 3 and by Figs. 3 and 4. For example, for a 200-Å-thick coating the irregular prismatic shape has up to 4.8 times more volume percent of coating material than the spheric shape with 1- $\mu\text{m}$ -size particles having up to 55.6 vol % (Table 3, Fig. 4). This, however, is not the maximum amount of possible enrichment. The upper limit is unknown but could approach 100% for the highly convoluted shapes that are common in lunar regolith materials. However, the spheric

Table 3. Relationship of Percent of Surface Coating Volume Versus Particle Width in  $\mu\text{m}$  for 200-Å-thick Surface Coating on 1- $\mu\text{m}$ -length Platic and Prismatic Shapes

Width, $\mu\text{m}$	% Volume	
	Plate	Prism
1.000	11.5264	11.5264
0.750	12.7552	13.9669
0.500	15.2128	18.7456
0.250	22.5856	32.2624
0.125	37.3312	55.6096

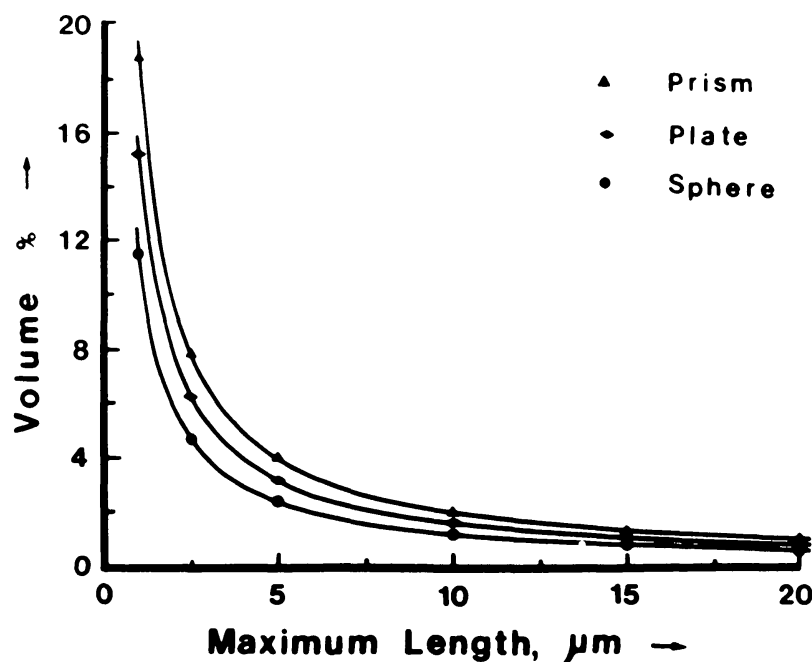


Figure 3. Relationship of volume percent of 200 Å surface coating versus maximum particle length in microns for particle shapes: sphere •, plate ◆, and prism ▲.

shape is one end member of a family of curves and is the worst case (Fig. 3) because, since a sphere has the minimum surface area to volume ratio of any shape, it has the minimum volume percent of surface coating of any shape.

### Percent Total Surface Coating Volume Relationships

The percentage of total surface coating volume is plotted versus grain diameter in Fig. 5, assuming a coating thickness of 200 Å. The calculation was done for a range of diameters from 1.0  $\mu\text{m}$  to 1000  $\mu\text{m}$ , although the plot is truncated at a diameter of 20  $\mu\text{m}$ . The curve in effect depicts the relation of cumulative volume enrichment as a function of grain size for a purely fictitious soil that has equal weight percents distributed in each of the calculated size bins. The figure shows that 90.9% of the total volume of surface coating occurs on the less than 20- $\mu\text{m}$ -size particles and that 82.1% occurs on the less than 10- $\mu\text{m}$ -size particles. Since real lunar soils often have higher weight percentages in the finer size fractions, a real cumulative fraction could be even more favorable for beneficiation of the lunar fines. Calculations using other coating thicknesses show similar trends.

## LUNAR REGOLITH FINES

Bustin *et al.* (1984) found that 63% of the hydrogen in Apollo 15 sample 15021, which has an average of 41 ppm hydrogen, was contained in the less than 20- $\mu\text{m}$ -size fraction, which made up 23 wt % of the sample; similar results were found by DesMarais *et al.* (1974). The data from sample 15021 (Bustin *et al.*, 1984) were recalculated and are listed in Table 4 along with the theoretical calculations for 200-Å-thick coated spheric and prismatic shapes. A comparison of the lunar data with results of the theoretical

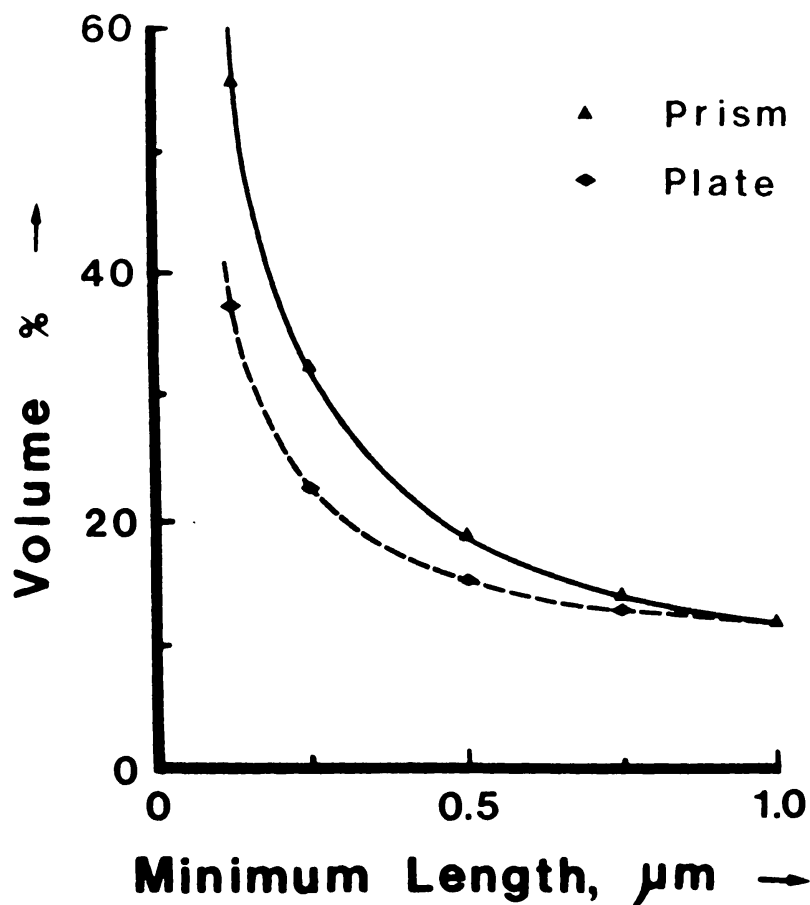


Figure 4. Relationship of volume percent of 200 Å surface coating on 1 μm maximum length particles versus shortest particle length in microns for particle shapes: plate ♦ and prism ▲

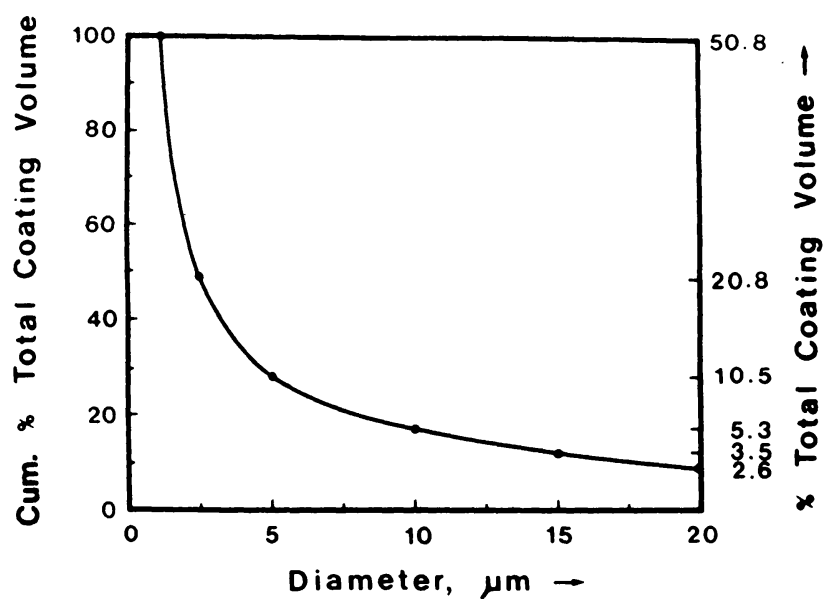


Figure 5. Relationship of cumulative percent of total coating volume versus diameter in microns for 200 Å surface coating on spheres.

Table 4. Relationship of Percent of Surface Coating Volume and Percent of Total Coating Volume to Particle Diameter and Particle Shape for 200-Å-thick Surface Coating

Average Particle Diameter, $\mu\text{m}$	Spheric % Vol.	% Coating	Prismatic* % Vol.	% Coating	% Hydrogen Content <sup>†</sup>
10.0	1.195	56.46	1.987	56.41	51.34
32.5	0.369	17.42	0.614	17.43	17.11
60.0	0.200	9.44	0.333	9.45	9.73
82.5	0.145	6.87	0.242	6.88	6.38
120.0	0.100	4.72	0.167	4.73	5.03
200.0	0.060	2.83	0.100	2.84	4.36
375.5	0.032	1.51	0.053	1.51	4.03
750.0	0.016	0.76	0.027	0.76	2.01
Total	2.117	100.01	3.523	100.00	99.99

\*Maximum length is twice the width.

<sup>†</sup>Calculated from  $\mu\text{g/g}$  data by Bustin *et al.* (1984) for Apollo regolith fines sample 15021.

calculations (Table 4, Fig. 6) reveals excellent similarity, but there is a slight decrease of hydrogen overall in the less than 100- $\mu\text{m}$ -size fractions and a significant enrichment of hydrogen in the greater than 82.5- $\mu\text{m}$ -size fractions.

The presence of constructional particles (McKay *et al.*, 1971; Carter, 1971), namely agglutinates (DesMarais *et al.*, 1974) and other dust-welded particles, and other non-spherical particle shapes, would increase the hydrogen content for those grain sizes containing these types of particles. Constructional particles result from the welding together

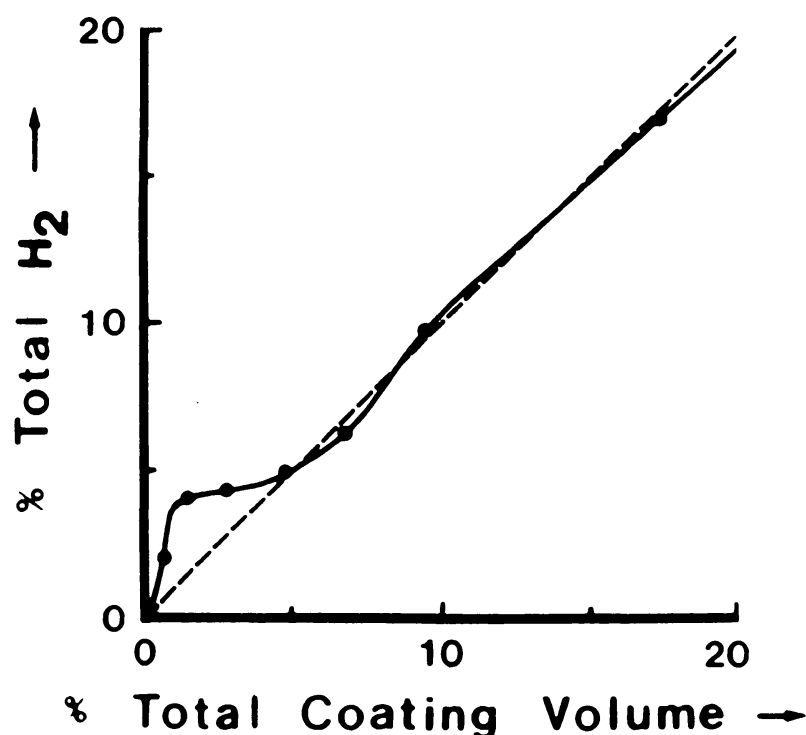


Figure 6. Relationship of percent of total hydrogen ( $\mu\text{m g/g}$ ) in grain size separates from Apollo 15 sample 15021 (listed in Table 4) versus percent of total surface coating volume on spheres. Dashed line is 1:1 relationship.

of regolith particles with liquid silicates during meteorite bombardment (McKay *et al.*, 1971) and should contain hydrogen values greater than those predicted by overall grain size alone. On the other hand, because a certain amount of the implanted hydrogen was released during the formation of the silicate liquid, as attested to by the presence of metallic iron spherules of non-splashed nature (e.g., Carter, 1973; Morris, 1980), an agglutinate or any other type of constructional particle, unless exposed to solar wind bombardment after formation, would have hydrogen contents lower than the theoretical predictions from total combined surface area of all the very fine size particles attached to or imbedded within its surface. However, unless there was loss of hydrogen during the formation of a constructional particle below the value predicted from overall grain size and there was no subsequent exposure of the particle to solar wind implantations, constructional particles would always contain excess hydrogen for a given grain size. This finding is supported by the research of DesMarais *et al.* (1974), who found agglutinates to be enriched in hydrogen relative to other particle types of similar grain size.

## BENEFICIATION

It is clear from Tables 2-4 and from Figs. 1-5 that, theoretically, the less than 20- $\mu\text{m}$ -size particles and certainly the less than 10- $\mu\text{m}$ -size particles contain significant surface coating volumes and will have to be physically separated to increase the surface coating materials to acceptable levels for exploitation. One possible way to accomplish this for lunar materials is to remove the material coarser than 100  $\mu\text{m}$  by vibratory screening and then separate the less than 20- $\mu\text{m}$ -size fraction by direct gaseous classification using turboscreening (Perry and Chilton, 1973). Finer size fractions, if necessary, can be made with modified cyclone techniques or possibly by electrostatic separation (Perry and Chilton, 1973; Agosto, 1984). Similar techniques are routinely used on Earth to classify particles to less than 2  $\mu\text{m}$  in diameter (Perry and Chilton, 1973). The optimum techniques can be determined best by experimentation with lunar simulants followed by testing of actual lunar materials under "lunar" conditions.

## EXTRACTION OF HYDROGEN

One aspect of the question of economics is how difficult and expensive it is to extract the hydrogen from the beneficiated (classified) lunar regolith fines. The hydrogen release pattern for a sample of Apollo 11 regolith fines (10086) (Gibson and Johnson, 1971) was recalculated and plotted in Figure 7. Their recalculated data reveals that approximately 81% of the hydrogen is released below 600°C using a heating rate of 4 °C/min. This means that only a moderate amount of thermal energy should be required to extract a significant portion of the hydrogen, especially if advantage is taken of daytime temperature on the lunar surface. Similar results are predicted from the thermal release patterns of samples of Apollo 12, 14, and 15 regolith fines (Gibson and Johnson, 1971; Gibson and Moore, 1972). A microwave technique has also been proposed to extract water from water-bearing lunar materials (Meek *et al.*, 1984).



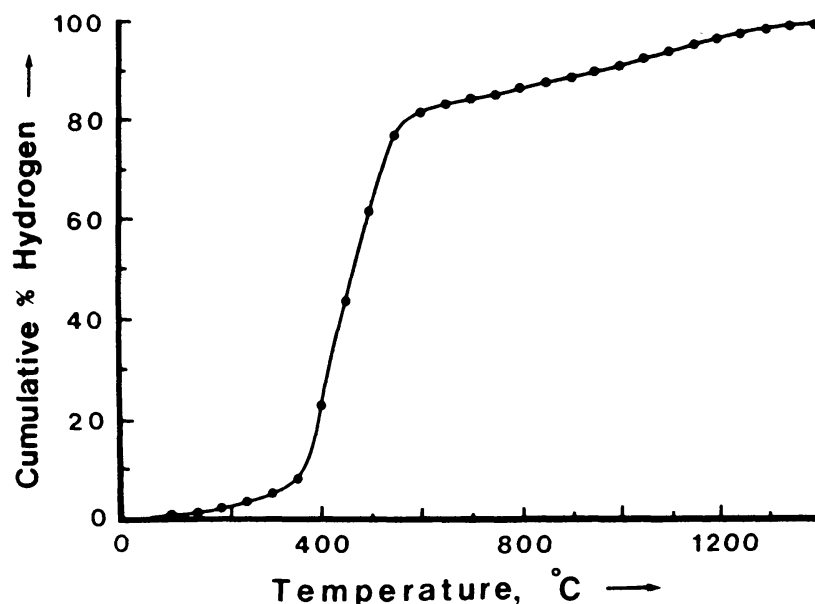


Figure 7. Relationship of cumulative percent of hydrogen versus temperature in degrees centigrade for Apollo 11 sample 10086,16. Data calculated from Gibson and Johnson (1971).

## DISCUSSIONS AND CONCLUSIONS

The theoretical rationale of lunar regolith fines as a primary source of hydrogen has been formulated and tested (Tables 2-4, Figs. 1-6). Similar arguments can be made for other elements implanted by solar wind bombardment or as surface condensates associated with volcanic eruptions (e.g., Chou *et al.*, 1975; Meyer *et al.*, 1975) or with meteorite impacts. The results listed in Tables 2-4 and displayed in Figs. 1-5 demonstrate that particles less than 20  $\mu\text{m}$  in size and especially particles less than 10  $\mu\text{m}$  in size have the theoretical capacity for significant concentration of hydrogen. Furthermore, the lunar regolith fines meet the basic requirement for beneficiation because a major portion of the hydrogen occurs in a relatively small part of the fines (Table 4). Particle shape is also a significant factor. For example, the prismatic shapes have up to 4.8 times more surface coating per maximum particle length than the spheric shape (Figs. 3 and 4).

The data shown in Figs. 1-6 result in mandatory size classification of the less than 20  $\mu\text{m}$  fraction. This should be accomplishable by a combination of vibratory screening followed by cyclone and/or possibly electrostatic separation. These methods are routinely used on Earth (Perry and Chilton, 1973) and should be adaptable to the lunar environment. However, the 1/6 g of the lunar environment may result in lower separation efficiencies. Only moderate amounts of thermal energy should be required to separate the hydrogen from the lunar regolith fines because approximately 81% of the solar wind trapped hydrogen is released by heating lunar regolith fines below 600°C (Fig. 7) (Gibson and Johnson, 1971; Gibson and Moore, 1972), or possibly, the hydrogen may be extracted with microwaves (Meek *et al.*, 1984).

Finally, the amount of material that would have to be processed to supply one ton of hydrogen is significant even when all efforts have been made to enhance the production

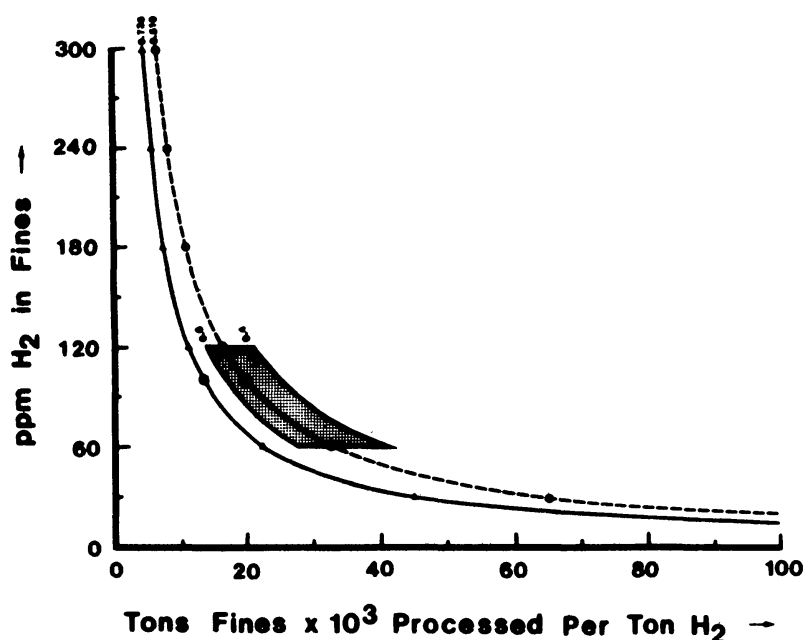


Figure 8. Relationship of number of tons of lunar regolith fines processed versus ppm hydrogen in the fines to yield one ton of hydrogen. (Number of tons of fines processed per ton of hydrogen =  $10^6/A \times 10^2/B \times 10^2/C$ ; where  $A$  = ppm hydrogen in fines;  $B$  = % of hydrogen in fraction heated; and  $C$  = % of hydrogen liberated during heating. Fractions are total recovery values.)

(Fig. 8). For example, assuming (a) an average of 100 ppm hydrogen (Table 1), (b) 90.9% of the hydrogen in the less than 20- $\mu$ m-size fraction (theoretical limit for spherical shapes), (c) 81% recovery of the hydrogen during heating of the concentrate (Fig. 7) and (d) 23 wt % of the lunar regolith fines in the less than 20- $\mu$ m-size fraction (Bustin *et al.*, 1984) a minimum of 13,582 tons of lunar regolith fines would have to be processed (Fig. 8) with 3,124 tons of concentrate heated yielding a theoretical recovery of 73.6% of the hydrogen. For 63% of the hydrogen in the less than 20- $\mu$ m-size fraction of Apollo 15 sample 15021 (Bustin *et al.*, 1984), a minimum of 19,596 tons of this type of lunar regolith fines would have to be processed (Fig. 8) with 4,507 tons of concentrate heated yielding a recovery of 51.0% of the hydrogen. The stippled area of Fig. 8 represents the most likely ranges for "typical" lunar fines. Although these tonnages are not high by terrestrial standards, they will probably limit early exploitation of the lunar regolith fines to hydrogen obtained as a by-product or co-product from the mining and processing for other elements or materials.

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## **HYDROGEN RECOVERY FROM EXTRATERRESTRIAL MATERIALS USING MICROWAVE ENERGY**

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The feasibility of recovering hydrogen from extraterrestrial materials (lunar and martian soils, asteroids) using microwave energy is presented. Reasons for harvesting, origins, and locations of hydrogen are reviewed. Problems of hydrogen recovery are discussed in terms of hydrogen release characteristics and microwave coupling to insulating materials. From results of studies of hydrogen diffusivities (oxides, glasses) and tritium release (oxides), as well as studies of microwave coupling to ilmenite, to basalt and to ceramic oxides, it is concluded that using microwave energy in hydrogen recovery from extraterrestrial materials could be the basis for a workable process of hydrogen recovery.

### **WHY HARVEST HYDROGEN FROM EXTRATERRESTRIAL MATERIALS?**

As manned space efforts proceed from the low Earth orbit activities of the latter part of this century toward colonization of the Moon, and ultimately Mars, in the 21st Century, propulsion and life support will place the greatest demands on technology and resources. Since the most advanced chemical propulsion systems employ hydrogen and oxygen as propellants, the availability of water becomes a significant issue. Although water is prevalent on the Earth and on solar system objects farther from the sun, the Moon exhibits no substantial evidence for it in free or bound (crystalline) states. However, schemes have been proposed to extract bound oxygen from lunar minerals and import hydrogen from the Earth in condensed forms. An alternate solution is to directly harvest hydrogen or water from extraterrestrial materials (lunar and martian soils, asteroids, etc.).

Lunar surface soils do contain hydrogen implanted from the solar wind, but in small concentrations. Much interest has been directed toward the possible presence of water ice within permanently shadowed craters at the lunar poles (Arnold, 1979), but without direct evidence it is best not to rely on its existence for current scenarios of Moon-base operations. Hydrogen occurs both as water ice and in hydrous minerals of carbonaceous chondrites of the asteroid belt. The farther outward we travel from Earth's moon, the better our chances of finding usable sources of hydrogen or water. Since water is vital to human use of space, we may find a useful trade-off between Earth-export plus local harvesting for lunar development and complete reliance on local harvesting for exploitation of Mars and the asteroids.

This paper deals primarily with the technical feasibility of recovering hydrogen from lunar soil using microwave energy as the heating source. Since hydrogen is abundant

on Mars and in certain types of asteroids, this technique could also be viable in hydrogen or water recovery from these sources.

## **ORIGINS OF HYDROGEN ON THE MOON, MARS, AND ASTEROIDS**

Hydrogen occurs almost entirely as soil-wind implanted hydrogen within the soil components. The fraction of this hydrogen that can occur as water is miniscule (probably  $10^{-3}$  to  $10^{-4}$ ; Blanford *et al.*, 1984). Thus the chances of obtaining primary water from lunar surface samples, as far as we know, are nil. Our knowledge of volatile-element systematics of lunar materials indicates that water is also internally deficient on the Moon, because the propellant gases for lunar volcanic gas-fountains were dominated by sulfur rather than by water and carbon-oxygen compounds as on Earth. In fact, the lowest hydrogen contents observed among lunar surface soils (0.2 ppm) occur in the gas-fountain orange glasses of Apollo 17 (Epstein and Taylor, 1973).

In contrast to Earth's moon, the hydrogen of Mars and of the carbonaceous chondrites appears to occur almost always as water ice or as water and structural OH within minerals. Moreover, the origins of this water are closely related to primordial formation or to atmospheric evolution (Mars). Primary water is directly available on Mars and on the carbonaceous chondrites.

## **LOCATIONS OF HYDROGEN ON THE MOON, MARS, AND ASTEROIDS**

Although lunar hydrogen is rare, the hydrogen that we know about is concentrated in lunar surface soils. In a depth profile obtained by studying the Apollo 15 deep drill core, Des Marais *et al.* (1974) found hydrogen concentrations of 124–184  $\mu\text{g/g}$  (or ppm) above 150 cm depth, and concentrations of 84–104  $\mu\text{g/g}$  below that depth. Bustin *et al.* (1984) report a range of 17–106  $\mu\text{g/g}$  hydrogen in 17 bulk soils representing all Apollo missions. Hydrogen contents in some bulk soils range up to almost 200  $\mu\text{g/g}$  (Epstein and Taylor, 1973; Des Marais *et al.*, 1974). A useful average concentration is 50  $\mu\text{g/g}$ , but this hydrogen is surface-correlated and is therefore most abundant in the finest soil fractions (Bustin *et al.*, 1984). However, our knowledge of how hydrogen is held by lunar soil particles is poor (Blanford *et al.*, 1984). For example, there is evidence that hydrogen abundance correlates with  $\text{TiO}_2$  content of a soil regardless of grain size (McKay, 1984).

How much hydrogen could we get from lunar surface soils? If an average surface soil abundance of 50  $\mu\text{g/g}$  is assumed, then a totally efficient extraction system would obtain about 50 grams of hydrogen, which could be converted to 0.45 liters of water, from each metric ton of soil processes. Obviously the extraction methods will not be totally efficient, and much of the surface area around a lunar base would have to be harvested to obtain appreciable amounts of water. Cost effectiveness of lunar hydrogen extraction will have to be weighed against the costs of hydrogen export from Earth in alternate forms such as condensed methane or ammonia (Friedlander, 1984).

Mars and the carbonaceous chondrites, compared to Earth's moon, are oases in space. In addition to having water ice at its polar caps, Mars may have permafrost extending to lower latitudes; the martian soil apparently contains hydrous minerals. Suggestions have been made that the hydrous mineralogy of martian soils at the Viking Lander sites may be dominated either by clay (Banin *et al.*, 1981) or by palagonite formed as an alteration product from the abundant martian basalts (Allen *et al.*, 1981). In either case the water can be extracted easily from these minerals by low-temperature heating. Carbonaceous chondrites contain serpentine-like hydrous silicates and perhaps some surface clays; spectral reflectance studies suggest that about 80% of the main-belt asteroids are carbonaceous chondrites (McCord and Gaffey, 1977) with water contents that may range from 1–20% (O'Leary, 1977). For extended human presence on Mars or among the asteroids, exportation of hydrogen from Earth may be necessary during initial exploration but should not be required for the development of permanent or semi-permanent bases.

## HYDROGEN RECOVERY

### Hydrogen Release Characteristics

For any type of hydrogen recovery process a major concern is the efficacy of the release of hydrogen from extraterrestrial materials, particularly from the lunar soils in which the concentration is so small. Several processes that appear to influence the rate of hydrogen release from a heated solid include the following:

1. Hydrogen solubility in the solid
2. Hydrogen diffusivity in the solid
3. Thermodynamic equilibria ( $H_2/H_2O$ )
4. Desorption kinetics
5. Grain boundary migration
6. Percolation outward through a packed bed

Each of these processes will have to be considered in some detail before the recovery of hydrogen from extraterrestrial material can be evaluated properly. Considerable data on hydrogen diffusivity in ceramics and glasses exists, much of it accumulated as part of the technology program for magnetic fusion energy. In that context, the studies were concerned with recovery of tritium ( $^3H$ ) bred in a lithium-containing blanket material from  $^6Li(n, \alpha)t$  and  $^7Li(n, n'\alpha)t$  reactions. If this tritium is bred in a solid material such as  $Li_2O$ ,  $LiAlO_2$ ,  $Li_4SiO_4$ , or  $LiZrO_3$ , it must be recovered with high efficiency. Studies of these recovery processes have generated much of the available tritium release data, which can readily be applied to hydrogen. Additional data are available on hydrogen and deuterium diffusivities in other ceramics and in quartz, pyrex, and borosilicate glass. A portion of these data is summarized in Table 1. In both pyrex and borosilicate glass the hydrogen species appear to migrate as an hydroxyl (OH) species. The hydrogen is released from these materials principally as water (Yamamoto, 1984).

Table 1. Selected Deuterium and Tritium Diffusion Coefficients

Diffusing Species	Material	$D_0$ ( $\text{cm}^2/\text{s}$ )	$Q$ , ( $\text{kJ/mole}$ ) C	Temp. Range	Reference
$D_2$	Pyrex glass	5.4	43.5	100–300	Laska <i>et al.</i> (1969)
$T_2$	Quartz	1–100	175	20–1000	Matzke (1967)
$T_2$	Fused silica	1–100	218	20–1000	Matzke (1967)
$T_2$	$\text{TiO}_2$	$1.8 \times 10^{-2}$	107	500–900	Cathcart <i>et al.</i> (1979)
$T_2$	$\text{Al}_2\text{O}_3$	$6.4 \times 10^{-5}$	132	300–1000	Cathcart <i>et al.</i> (1979)
$T_2$	BeO	$1.3 \times 10^{-6}$	129	300–1200	Fowler <i>et al.</i> (1977)

$$D = D_0 \exp (-Q/RT)$$

Tritium extraction from irradiated metallic aluminum–lithium alloys has been a familiar process since the 1950s. The technique involves heating the irradiated targets in a retort to drive off the tritium. Studies have also been conducted on extraction of tritium from irradiated lithium oxide, lithium aluminate, and lithium silicate targets. In laboratory experiments, ~95% of all tritium is recovered when the pre-irradiated samples are held at 800°C for 4 hours (Johnson *et al.*, 1976). Progressively greater recovery is obtained at 800°C with extended anneals at this temperature. Anneals of 10–15 minutes at 1300°C appear to result in almost complete tritium recovery from the aluminate and silicate targets.

The main fraction of the gases evolved from the target capsules during laboratory extractions is condensible ( $\text{HTO}$  and  $\text{T}_2\text{O}$ ). Other experimenters have likewise observed that at temperatures above 600°C a major fraction of the released tritium is indeed in the condensible form. In addition to the silicate and aluminate, a limited number of tests were performed with petalite ( $\text{Li}_2\text{O}-\text{Al}_2\text{O}_3-8\text{SiO}_2$ ). Tritium release characteristics appeared to be quite similar for both aluminate and petalite.

Consideration of the available data on hydrogen, deuterium, and tritium diffusivities and release characteristics in glasses and ceramics leads one to conclude that if extraterrestrial materials can be heated to 600°–1000°C, there is reasonable probability for release of the hydrogen from these materials. Whether the hydrogen is recovered as gaseous  $\text{H}_2$  or as the oxide ( $\text{H}_2\text{O}$ ) is uncertain. Some insight into the relative probabilities of these two alternatives is doubtless possible from thermochemical techniques using complex chemical equilibrium calculations. Such calculations would predict the equilibrium composition of gas released from lunar soils containing implanted hydrogen when heated.

### Application of Microwave Heating in Hydrogen Recovery

The use of microwave energy would offer a new, very efficient method of heating lunar materials. Not only could extraterrestrial materials be heated with less energy than with conventional methods, but the heating would be accomplished in a controlled manner

and in much less time than with a method such as solar heating. This is due in part to the ability to control field intensities, heating rates, and maximum heating temperature during microwave heating. Also, the thermal conductivity of the lunar soil surface is estimated by Langseth *et al.* (1972) to be  $60 \mu\text{W}/\text{cm-K}$ , which is two to three orders of magnitude less than that for oxides such as alumina. Thus, the use of focused solar radiation to quickly heat the lunar-soil volumes appears impractical. The top centimeter or so of lunar soil would strongly impede the diffusion of heat, possibly fusing the soil and preventing release of subsurface hydrogen. In contrast, microwaves tend to heat a soil volume uniformly because their low attenuation results in a fairly uniform released-energy density.

As a specific example, microwave generators could be attached to a mobile vehicle, allowing *in-situ* heating of the upper regolith layer. If the hydrogen was released principally as  $\text{H}_2\text{O}$ , the water vapor could be condensed on a cold platter immediately above the surface. If only hydrogen is released, it will evolve from the lunar surface in molecular form, presenting a significant collection problem. Since such a system would eliminate the extensive mining and hauling associated with a retorting process, it will be important to study evolved lunar soil volatiles and to investigate innovative collection techniques for molecular hydrogen in the lunar environment. An important aspect of the recovery process is the coupling of microwave energy in insulating materials such as basalts, ilmenite, and feldspars, which are widely present on the lunar surface and likely to be found on Mars. Microwave energy at 2.45 GHz is known to couple to such minerals as well as activate dielectric loss mechanisms such as ion migration, ionic vibration, and impurities. As the temperature increases from approximately  $25^\circ\text{C}$ , thermally activated processes (such as thermal activation of electrons from valence to conduction bands) that affect conductivity will also impact overall loss characteristics.

Since the early works by Von Hippel (1954a,b), there has been interest in how microwave energy couples into oxide materials. Perhaps the best work done in this area has been by Tinga (1969, 1970), by Tinga and Edwards (1968), by Tinga and Nelson (1973), and by Tinga and Voss (1969). Table 2 lists some materials that have been thermally processed along with observed heating rates for three glasses using microwave energy. All work to date has been accomplished at the two allowed industrial frequency bands (915 MHz and 2.45 GHz).

Table 2. Some Materials Heated by Microwave Energy at Los Alamos National Laboratory

Material	Melting Temperature	Observed Heating Rate
OI - 1756 Glass	$462^\circ\text{C}$	12,000 C/hr
OI - 0038 Glass	$735^\circ\text{C}$	200,000 C/hr
OI - 1613 Glass	$1450^\circ\text{C}$	33,000 C/hr
Alkali Basalt	$1200^\circ\text{C}$	—
$\text{Al}_2\text{O}_3$	$2059^\circ\text{C}$	—



High absorptivity of microwave energy is obviously desirable if processing by use of this energy source is to be feasible. Energy absorption of a plane wave in a dielectric material increases explicitly with frequency and also with the product of loss tangent and dielectric constant (Frost, 1985). Loss tangent shows much more variability with frequency than does the dielectric constant and is therefore the focus of most attention when considering dielectric losses. Impurities and defects play an important role in absorption and thus in the coupling of energy to the material in question. At a given temperature and frequency, there is an "intrinsic" loss characteristic of the chemical formulation and crystal structure of the material. As a rule, loss tangent is higher than this intrinsic value due to the presence of chemical and structural defects. Whether energy absorption is due to oscillation of a charged species (e.g., ion or electron) or to scattering from grain or interphase boundaries, higher frequencies such as those mentioned earlier may be expected to couple more effectively into loss processes associated with defects of quite small scales such as might exist in lunar soils.

Mobile defects and ions that enhance the conductivity of oxide materials are known to couple to microwave energy at the 2.45 GHz and 915 MHz frequencies, increasing energy absorption. For example, beta alumina, which contains 11% sodium by weight, can be heated from room temperature to its sintering temperature (1850°C) in just a few seconds when placed in a 2.45 GHz microwave field of 400 watts power. Materials such as  $\text{Cu}_{2x}\text{O}$ , ZnO, and  $\text{ZrO}_2$  also should couple efficiently because they are defect-controlled semiconductors. In order to accomplish heating of traditional oxide materials such as alpha alumina, we incorporate materials that do couple at 2.45 GHz radiation such as glycerol or sodium nitrate. These materials cause the oxide to heat to a few hundred degrees Celsius, after which the oxide itself will then couple because the loss tangent will increase sufficiently with temperature to allow the material to absorb electromagnetic energy efficiently.

Initial work with terrestrial alkali basalt has shown that in order to heat this material, a coupling agent is needed. Recently we have shown the ability to heat an ilmenite-rich rock to its melting temperature using 2.45 GHz microwave energy without a coupling agent. Since ilmenite is present on the lunar surface in abundant quantities, it could act as a coupling agent to allow the initial heating of those lunar materials that may not couple at ambient temperature.

Also, it is known that most lunar soil, at least down to a depth of one foot, contains about  $10^6$  cosmic or solar fossil particle tracks per cc. The defects induced into this soil over millions of years of exposure to solar wind and cosmic ray events should enhance the loss tangent of this material and perhaps allow lunar materials to be heated without the use of coupling agents in a microwave field.

One variable that has not been investigated but that has much control over whether a material couples or not, is the field intensity (E,H). On the Moon, heating will be in a vacuum (high dielectric breakdown strength) where very high fields can be used. Consequently, materials that would not couple on Earth may heat very easily and quickly on the Moon.

## CONCLUSIONS AND RECOMMENDATIONS

This paper has discussed hydrogen recovery from extraterrestrial materials in general and in particular from lunar soils, using microwave energy. Results have been presented that are relevant to the prospects for microwave processing. One can conclude that if enough effort is invested in further study, this process could produce techniques in which energy can be used in a viable hydrogen recovery process.

Much work remains to fully characterize some of the phenomena observed to date with microwave-heated oxide and composite materials. For example, characterization of the species in which hydrogen is released from oxides will require chemical equilibrium calculations, diffusion modeling, study of reaction and sintering kinetics, and characterization of microwave coupling processes. Also, in order to be certain that lunar materials can be heated in a microwave environment, some small amount of well characterized lunar material should be thermally processed in such an environment and fully restudied after processing.

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# MICROBIAL EXTRACTION OF HYDROGEN FROM LUNAR DUST

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If molecular hydrogen in lunar dust can be made available to the hydrogenases of bacteria, then several microbial pathways exist for the potential liberation of hydrogen, carbon dioxide, and methane using relatively simple apparatus. Intermediate products include microbial biomass and short chain organic acids such as acetate. The hydrogen could be harvested, and carbon dioxide, phosphate, nitrogen, and trace nutrients could be recycled. All reactions suggested in this paper, or similar ones, have been demonstrated on Earth, with the exception of the initial utilization of hydrogen from lunar fines by bacterial hydrogenases. However, it is possible to test these reactions on extant samples of lunar fines. If potentially toxic elements in lunar soils present problems for such processes, bacterial tolerance can be induced by plasmid transfer or by selection among cells subjected to increasing levels of these elements.

## INTRODUCTION

A future permanent human colony on the Moon should be as independent from Earth as possible. Rocket fuel, for instance, should be produced from lunar resources to reduce the high costs of transportation from Earth (Duke *et al.*, 1985). Detailed analyses of samples brought back during the Apollo missions have shown that lunar regolith fines contain, mostly in their 20  $\mu\text{m}$  size fraction, substantial amounts of extractable hydrogen (Bustin *et al.*, 1984). The  $\text{H}_2$  concentrations varied from 50–220 ppm; average values given were 100–150 ppm, or 100–150  $\mu\text{g/g}$  regolith fines. This hydrogen can be released by heat. The economic viability of the lunar base could be greatly improved if the hydrogen held in the lunar regolith fines could be harvested by bacteria, which would be needed anyway to aid in the recycling of human organic wastes.

Bacteria offer particularly versatile mechanisms of harvesting lunar hydrogen as they contain self-correcting features such as multiple homeostatic abilities to regulate their microenvironment. They show genetic versatility that allows them to compensate for a wide variety of conditions. Bacteria can be thought of as self-replicating catalysts with the power to adapt to their environment.

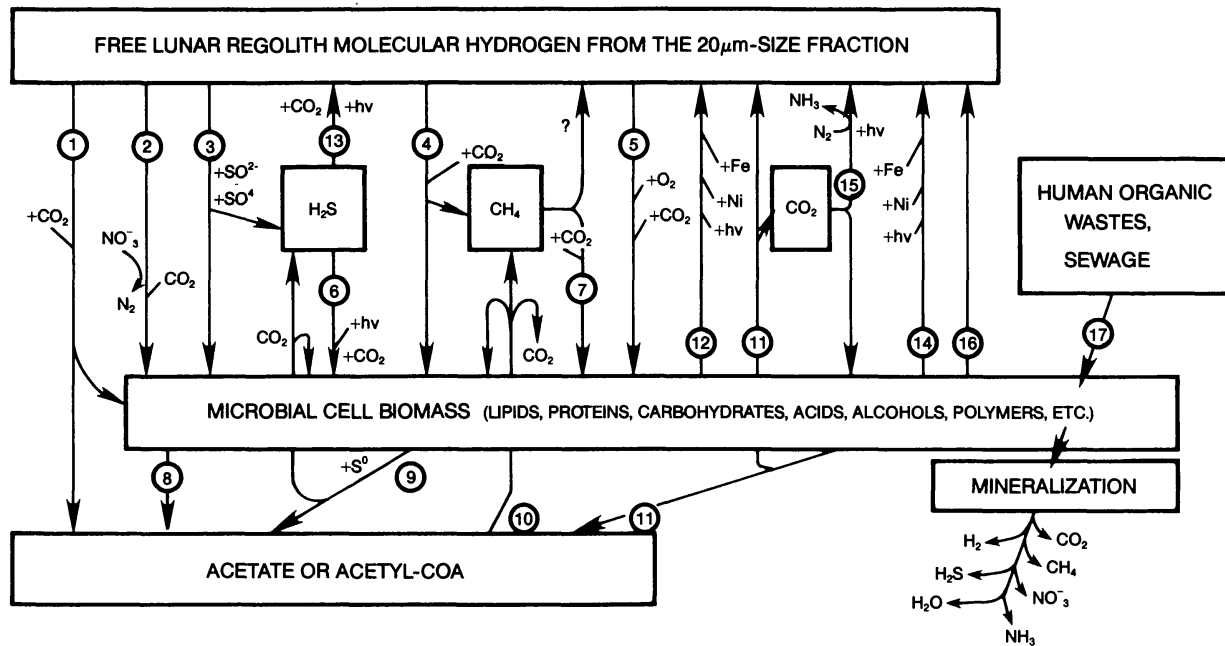


Figure 1. Possible reactions by which molecular hydrogen could be harvested from lunar dust with microbial activities. 1-Acetobacterium woodii, A. carbinolicum, Methanobacterium thermoautotrophicum, Clostridium thermoaceticum, Sporomusa sphaeroides. 2-Paracoccus denitrificans. 3-Desulfovibrio vulgaris; Campylobacter sp. 4-Methanobacterium spp. 5-see Table 2. 6-Chromatiaceae, chlorobacteria. 7-Rhodospseudomonas gelatinosa. 8-Acetobacterium carbinolicum, Ilyobacter polytrophus. 9-Thermococcus celer, Desulfurococcus mucosus. 10-Methanosarcina barkeri. 11-Acetomicrobium flavidum, Acidaminobacter hydrogenoformans. 12-Purple bacteria. 13-Chromatiaceae. 14-Rhodospirillaceae. 15-Cyanobacteria. 16-Escherichia coli. 17-Anaerobic sewage fermentations; aerobic treatment.

We propose that especially anaerobic bacteria could serve as a mechanism for harvesting the lunar hydrogen. The microbial biomass and organic compounds into which the hydrogen would be fixed initially could then be converted to molecular hydrogen by employing special bacterial strains or consortia and special conditions (Fig. 1). With the exception of the initial reaction whereby hydrogen is released from lunar dust, and whereby the bacteria are subjected to possibly toxic regolith mineral components, all subsequent steps are well established in microbial systems existing on Earth.

In a first step, anaerobic bacteria utilizing the hydrogen could create organic acids, CO<sub>2</sub>, H<sub>2</sub>S, or CH<sub>4</sub>, as well as biomass. As in terrestrial sewage plants, anaerobic digesters could produce more methane from this biomass. Immobilized anaerobic and phototrophic bacteria in illuminated reaction chambers could generate CO<sub>2</sub> and H<sub>2</sub>. After harvesting the hydrogen, the CO<sub>2</sub>, phosphate, nitrogen, and trace nutrients could be recycled.

At a later stage, with oxygen available from iron-oxide-containing minerals, aerobic processes could generate water from the bacterial oxidation of hydrogen. Also, aerobic methane-oxidizing bacteria would convert CH<sub>4</sub> and CO<sub>2</sub> into water and biomass. The

water could be dissociated into oxygen and hydrogen. Aerobic hydrogen release by N<sub>2</sub>-fixing cyanobacteria (blue-green algae) would be another possibility that could be explored.

## ENVIRONMENTAL CONDITIONS NEEDED FOR BACTERIAL CULTIVATION

The conditions required are relatively simple to engineer. The necessary bacterial cultures can be brought from Earth in the form of either lyophilized or deeply-frozen strains. Lyophilization (freeze-drying) is performed by exposing well-grown cultures to very high vacuum in the cold. The dried cells are sealed in glass vials while attached to the vacuum line. Lyophilized cells are extremely resistant to noxia such as temperature changes, *etc.* As they do not grow in this dry state, they cannot express mutations occurring occasionally due to radiation. Transportation from Earth to the Moon in a spaceship would, therefore, be possible. No measurable losses of viability were noted with *Hydrogenomonas eutropha* Z1 and *Escherichia coli* K 12 when flown aboard the Russian satellite Cosmos 368 or aboard the automatic lunar station Zond 8 (Taylor, 1974).

The pressure would be optimized by engineering studies and would most likely not exceed several Earth atmospheres. Fermenters for all of these reactions could be opaque, but in the case of photodissociation of organics by purple or green bacteria, some protection from high light intensities and from short wavelength ultraviolet light would be needed. Nitrogen, sulfur, and possibly phosphorus, as well as most trace minerals necessary for the functioning of specific enzymes, would be available from lunar regolith fines. The relative concentrations of these might have to be altered.

In the first stage of an optimal system, the lunar fines would be added to anaerobic fermenters and the hydrogen harvested in the form of bacterial biomass and fermentation products. The second stage would utilize reactors in which phototrophic anaerobes (purple or green bacteria) could be immobilized on special surfaces and generate H<sub>2</sub> in the light. The reactor fluid and other gases containing CO<sub>2</sub>, as well as phosphate, sulfur, nitrogen, trace minerals, vitamins, and water would be recycled, partly by microbial processes ("mineralization"), partly chemically.

All organic wastes and sewage generated by the lunar population would be collected and fermented much as in anaerobic sewage treatment facilities on Earth, to provide additional products for the photoreactors.

In the third stage, with oxygen available from iron-oxide-rich lunar minerals, aerobic autotrophic bacteria or unicellular green algae would be employed as well. Especially selected or engineered aerobic strains with high hydrogenase activity would be able to tolerate stress conditions of the lunar environment. These aerobic organisms could extract H<sub>2</sub> from the regolith in explosion-proof fermenters; their Knallgas reaction would form water and biomass.

Some recently discovered archaebacteria (*Sulfolobus ambivalens*) grow either anaerobically with H<sub>2</sub>, S<sup>0</sup>, and CO<sub>2</sub> to form H<sub>2</sub>S and cell biomass or aerobically with

$S^0$ ,  $O_2$ , and  $CO_2$  to form sulfate (Zillig *et al.*, 1985). To work with bacterial strains of great metabolic versatility would certainly be advantageous.

## HYDROGEN UTILIZATION

The initial reaction, which is critical to the whole operation, requires the release of hydrogen from the lunar soil. Although pretreatments may be required, the essential interaction is between the enzyme hydrogenase in the bacteria and the lunar hydrogen. Well-known hydrogenases for the initial anaerobic step occur in two candidate microbes: the sulfate-reducing and the methanogenic bacteria. Sulfate-reducing bacteria form biomass and  $H_2S$  from  $SO_4^{2-}$ ,  $H_2$ , and  $CO_2$  (plus trace minerals and possibly some vitamins). The Michaelis-Menten constant  $K_m$ , for growth with hydrogen-limiting conditions for a number of  $SO_4$  reducers, is  $2 - 4 \times 10^{-6}$  Molar (Robinson and Tiedje, 1984). (The  $K_m$  is the concentration of  $H_2$  at which growth is at the half maximal rate.) Methanogenic bacteria that form  $CH_4$  and biomass from  $H_2$  and  $CO_2$  have  $K_m$  values of  $6 - 7 \times 10^{-6}$  Molar hydrogen. Sulfate reducers thus can outcompete the methanogens for hydrogen. The  $H_2S$  generated from sulfate-reducing bacteria may pose corrosion problems in the reactors, as it does on Earth, but, of the organisms that have been well studied, these have the highest known affinity for hydrogen.

Table 1. Anaerobic Bacterial Hydrogen Consumption Yielding Various Reduced Compounds and Cell Biomass (CBM)

Organisms	Reactions	References
<i>Paracoccus denitrificans</i>	$5H_2 + 2H^+ + 2NO_3^- \longrightarrow N_2 + 6H_2O + CBM$	Schlegel and Schneider (1978)
<i>Campylobacter</i> sp.	$H_2 + S^0$ (or $S_2O_3^{2-}$ , or $SO_3^-$ ) $\longrightarrow HS^- + H + CBM$	Laanbroek <i>et al.</i> 1978
<i>Desulfovibrio vulgaris</i> (strain Marburg)	$4H_2 + H^+ + SO_4^{2-} \longrightarrow HS^- + 4H_2O + CBM$	Schlegel and Schneider (1978)
<i>Methanobacterium</i> sp.	$4H_2 + CO_2 \longrightarrow CH_4 + 2H_2O + CBM$	Mah and Smith (1981)
<i>Methanosphaera stadtmaniae</i>	$H_2 + CH_3OH \longrightarrow CH_4 + H_2O + CBM$	Miller and Wolin (1985)
<i>Vibrio succinogenes</i>	$H_2 + \text{fumarate}^{2-} + \longrightarrow \text{succinate}^{2-} + CBM$	Schlegel and Schneider (1978)
<i>Acetobacterium carbinolicum</i>	$4H_2 + 2HCO_3^- + H^+ \longrightarrow \text{acetate} + 4H_2O + CBM$	Eichler and Schink (1984)
<i>Acetobacterium woodii</i>	$H_2 + CO_2 \longrightarrow \text{acetate} + CBM$	Ragsdale and Ljungdahl (1984)
<i>Methanobacterium thermoautotrophicum</i>	$8H^+ + 2CO_2 \longrightarrow \text{acetyl-CoA (acetate)} + CBM$	Stupperich and Fuchs (1984)
<i>Sporomusa sphaeroides</i>	$28H^+ + 7CO_2 \longrightarrow \text{acetate} + CBM$	Möller <i>et al.</i> (1984)
<i>Clostridium thermoaceticum</i>	$H_2 + CO_2 + \text{CoASH} + \text{methyl-THF} \longrightarrow \text{acetyl-CoA} + \text{THF} + CBM$	Pezachka and Wood (1984)

Since the reaction of the lunar hydrogen with these bacteria is the untested portion of our harvesting scheme, we propose that a large number of different bacterial types be tested. Suggested anaerobic bacteria are listed in Table 1. Products of hydrogen fixation are water, sulfide, methane, succinate or acetate, and cell biomass.

It is also possible to create microbial biomass and water from hydrogen aerobically, once oxygen becomes available either as an import or as a locally produced commodity. The process requires digesters with vigorous aeration, the engineering of which is well known. A selected list of aerobic bacterial candidates is given in Table 2. The list contains representatives of very different physiological groups. Some aerobic H<sub>2</sub> bacteria excrete large amounts (up to 5g/l) of organic compounds, especially when NH<sub>4</sub><sup>+</sup> and oxygen are limiting (Vollbrecht *et al.*, 1978).

Table 2. Aerobic Bacteria with the Ability for Hydrogen Consumption and the Formation of Water and Cell Biomass (CBM)

Organisms	References	Organisms	References
<i>Pseudomonas saccharophila</i>	Schlegel and Schneider (1978)	<i>Corynebacterium</i> sp.	Jochens and Hirsch, (personal communication (1985)
<i>Alcaligenes eutropha</i>	Schlegel and Schneider (1978)	<i>Azospirillum lipoferum</i>	Malik and Schlegel (1981)
<i>Seliberia</i> sp.	Schlegel and Schneider (1978)	<i>Derria gummosa</i>	Malik and Schlegel (1981)
<i>Comamonas</i> sp.	Schlegel and Schneider (1978)	<i>Rhizobium japonicum</i>	Malik and Schlegel (1981)
<i>Hydrogenobacter</i> sp.	Schlegel and Schneider (1978)	<i>Microcycilus aquaticus</i>	Malik and Schlegel (1981)
<i>Xanthomonas autotrophicus</i>	Schlegel and Schneider (1978)	<i>Microcycilus eburneus</i>	Malik and Schlegel (1981)
<i>Rhizobium leguminosarum</i>	Schlegel and Schneider (1978)	<i>Renobacter vacuolatum</i>	Malik and Schlegel (1981)
<i>Mycobacterium phlei</i>	Hirsch (1961)	<i>Rhodopseudomonas capsulata</i> (dark)	Gogotov (1984)
<i>Nocardia opaca</i>	Hirsch (1961)	<i>Rhodospirillum rubrum</i> (dark-grown)	Gogotov (1984)
<i>Nocardia hydrocarbonoxides</i>	Hirsch (1961)	<i>Rhodopseudomonas acidophila</i> (dark)	Schlegel and Schneider (1978)
<i>N. petroleophila</i>	Hirsch (1961)	<i>Gloeobacter</i> sp. 7421	Howarth and Codd (1985)
<i>N. autotrophica</i>	Hirsch (1961)	<i>Synechococcus</i> sp. 6307	Howarth and Codd (1985)
<i>Nocardia</i> sp. from Antarctic rocks	Hirsch (personal communication, 1985)		



## METHANE UTILIZATION

Organic wastes from human activities and the combined cellular biomasses produced anaerobically or aerobically would be fermented in anaerobic digesters resulting in the production of methane, carbon dioxide, and possibly some short chain organic acids, using technology already well studied on Earth.

Numerous species of aerobic bacteria are known to oxidize methane (Whittenbury *et al.*, 1970): *Methylobomonas*, *Methylycoccus*, *Methylosinus* spp. From the anaerobic fermenters they could harvest methane, a particularly attractive product as it is easily handled. These bacteria form water and cell biomass in the presence of oxygen. They can also fix nitrogen if the level of oxygen is low. The water produced would be harvested and electrochemically dissociated into hydrogen and oxygen. It could also be dissociated biologically by using immobilized cyanobacteria (Kayano, *et al.*, 1981).

## BIOLOGICAL RELEASE OF HYDROGEN

The terminal stage of the process involves the production of  $H_2$ . Numerous phototrophic bacteria can be grown under conditions where hydrogen is released in the light. We have summarized some of these reactions in Table 3; they are examples of a large number

Table 3. Microbial Hydrogen Evolution from Various Substrates

Organisms	Reactions	References
<i>Rhodopseudomonas</i> sp., <i>Thiocapsa</i> sp., <i>Chromatium</i> sp., <i>Thiocystis</i> sp.	lactate, acetate, ethanol, $S^0$ , or $S^{2-}$ $\xrightarrow[\text{anaerobic}]{\text{light}}$ $\longrightarrow H_2 + \text{oxidized substrate} + \text{CBM}$	Gogotov (1984); Matheron and Baulaigue (1983)
<i>Rhodopseudomonas</i> sp. (strain MPBE 2271)	malate $\xrightarrow[\text{anaerobic}]{\text{light}}$ $H_2 + CO_2 + \text{CBM}$	Mitsui <i>et al.</i> (1983)
<i>Escherichia coli</i>	sugars, pyruvate, or formate $\longrightarrow X + 2H^+ + \text{CBM} + \longrightarrow H_2$	Adams <i>et al.</i> (1981)
<i>Acetomicrobium flavidum</i>	hexose + $2H_2O \xrightarrow{58^\circ C} 4H_2 + 2CO_2 + 2 \text{ acetate} + \text{CBM}$	Soutschek <i>et al.</i> (1984)
<i>Clostridium</i> sp.	pyruvate or molasses $\xrightarrow{\text{ferredoxin}}$ $H_2 + \text{CBM}$	Schlegel and Schneider (1978); Suzuki <i>et al.</i> (1983)
<i>Rhodopseudomonas</i> <i>gelatinosa</i>	$CH_4 + CO_2 \xrightarrow[\text{anaerobic}]{\text{light}}$ $\text{CBM} + H_2?$	Wertlieb and Vishniac (1967)
<i>Nostoc</i> sp. <i>Gloeobacter</i> sp. 7421	$N_2 + CO_2 \xrightarrow[\text{air}]{\text{light}}$ $H_2 + H_2O + O_2 + NH_3 + \text{CBM}$	Kerfin and Böger (1982), Howarth and Codd (1985)
<i>Synechococcus</i> sp.	$N_2 + CO_2 \xrightarrow[\text{air}]{\text{light}}$ $H_2 + H_2O + O_2 + NH_3 + \text{CBM}$	Mitsui <i>et al.</i> (1983)
<i>Acetaminobacter</i> <i>hydrogenoformans</i>	glutamate $\xrightarrow[\text{anaerobic}]{} H_2 + \text{acetate} + CO_2 + \text{propionate}$ or formate	Stams and Hansen (1984)

of similar reactions known to occur in bacteria and even in green algae. In the latter case, strains of *Chlorella pyrenoidosa*, *C. vulgaris*, or *Selenastrum gracile* actively produce hydrogenase approximately four hours after onset of anaerobiosis (Kessler and Maifahrth, 1960). The hydrogen production of immobilized cells of *Clostridium butyricum* has been studied quantitatively by Suzuki *et al.* (1983). Three kilograms of wet cells produced, from molasses, 400–800 ml/min<sup>-1</sup> H<sub>2</sub> continuously over a 2-month period. An engineering analysis of photosynthetic hydrogen production systems was carried out by Herlevich *et al.* (1983), who calculated the cost of production.

Possibly a reisolation of the *Rhodopseudomonas* observed by Wertlieb and Vishniac (1967) would be attempted. This bacterium oxidized CH<sub>4</sub> with CO<sub>2</sub> in light, anaerobically, with the formation of cell biomass. Under nitrogen-limiting conditions, it might be possible to induce the organism to release hydrogen in the light. Of course, it is also possible to develop an abiotic process to photochemically release hydrogen from methane.

The metabolic versatility of non-sulfur purple bacteria (*Rhodospirillaceae*) could be advantageous. These organisms readily utilize hydrogen as an electron donor for photoautotrophic growth (Stanier *et al.*, 1976). The mechanisms used by these anaerobes resemble those of aerobic hydrogen bacteria (Schneider and Schlegel, 1977), but aerobic hydrogen consumption requires the absence of light. In the presence of light, under anaerobic conditions, nitrogen limitation induces N<sub>2</sub>-fixation, and their nitrogenase releases H<sub>2</sub>.

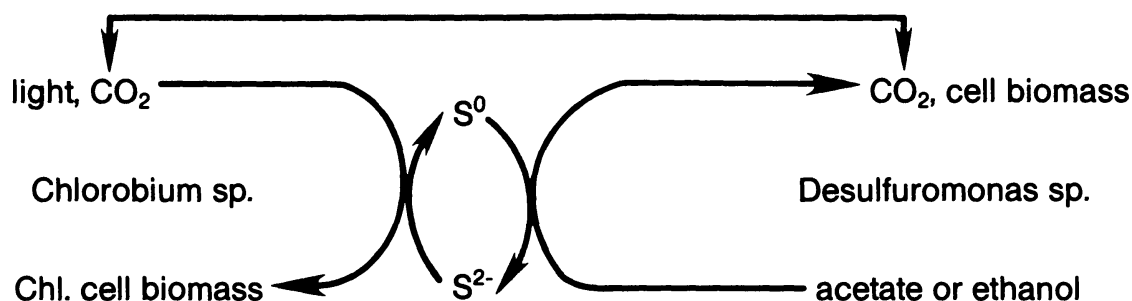
## INTERMEDIATE REACTIONS YIELDING ACETATE

Conversion of various organic excretion products to acetate may become necessary. Acetate is a possible precursor for methane (Mah *et al.*, 1978) or photosynthetic hydrogen evolution (Table 3). *Ilyobacter polytrophus* ferments 3-hydroxybutyrate, crotonate, pyruvate, citrate, glucose, fructose, malate, or fumarate, to form acetate as well as some formate, propionate, and butyrate (Stieb and Schink, 1984). Another acetogenic bacterium is *Acetobacterium carbinolicum*, which ferments formate, methanol, aliphatic alcohols, 1,2-diols, 2,3-butanediol, acetoin, glycerol, lactate, pyruvate, or hexoses (Eichler and Schink, 1984). Glutamate is converted to acetate, CO<sub>2</sub>, formate, propionate, and H<sub>2</sub> by *Acidaminobacter hydrogeniformans* (Stams and Hansen, 1984). Such versatile bacteria would be preferred to fermentation specialists that could utilize only few compounds.

## FORMATION OF REDUCED SULFUR COMPOUNDS

Hydrogen fixation by sulfate-reducing bacteria has already been mentioned (Table 1). Several newly discovered anaerobic bacteria utilize organic compounds as hydrogen donors and S<sup>0</sup> as the hydrogen acceptor. *Desulfurococcus mucosus* and *D. mobilis* grow with yeast extract or casein as carbon and hydrogen source and need S<sup>0</sup> as the acceptor, producing sulfide and CO<sub>2</sub> (Zillig *et al.*, 1982). These organisms are extremely thermophilic, requiring 85°C for optimal growth. Similar reactions with peptides are carried out by *Thermococcus celer*, a moderately halophilic thermophile that requires 88°C and 40 g/l of NaCl for growth (Zillig *et al.*, 1983). Finally, sulfide production from S<sup>0</sup> and acetate

has been described for *Desulfuromonas acetoxidans*, an anaerobic bacterium that lives syntrophically with green phototrophic sulfur bacteria of the genus *Chlorobium* (Pfennig and Biebl, 1976):



The sulfide-hydrogen that accumulates in such reactions would be transferred into purple bacterial biomass (*Chromatiaceae*) through their anaerobic photosynthesis. Under aerobic conditions, the sulfide-reducing equivalents could be harvested by sulfide-oxidizing bacteria such as *Beggiatoa* spp., *Thiothrix* spp., *Thiobacillus* spp., etc.

## POSSIBLE TOXICITY OF LUNAR REGOLITH COMPONENTS

In the first reactions proposed here, the H<sub>2</sub>-utilizing bacteria and their enzymes will be in direct contact with the lunar regolith fines. Analyses of Apollo soil samples taken from mare, highland, or basin ejecta show relatively high concentrations of some potentially toxic elements (Table 4). Although some of the elements occur in higher concentrations

Table 4. Concentration Ranges of Selected Lunar Soil Elements with Potential Toxicity to Microorganisms

Element	Concentration Range in Lunar Soil ( $\mu\text{g/g}$ soil)
As	0.01–0.41
Ba	85.7–767.5
Be	1.2–5.5
Cd	0.03–0.8
Cr	700.0–3600.0
Cu	6.4–31.0
F	37.0–278.0
Mn	500.0–1900.0
Ni	131.0–345.0
Pb	0.8–10.0
Se	0.03–0.39
Sr	104.2–234.0
U	0.26–3.48
Zn	6.3–49.0

From Freitas and Gilbreath (1982)

on Earth, their combined presence could be detrimental to normal terrestrial bacteria. Lunar samples containing high chromium and manganese may have to be tested for such toxic effects.

Microorganisms can develop extreme tolerances to heavy metals and some rare elements (Tyler, 1981). The detoxification can be accomplished by several different methods. Cell walls of bacilli and filamentous fungi can sequester heavy metals. Chitin or chitosan of *Rhizopus arrhizus* can sequester uranium or thorium at amounts of up to 20% of the organism's dry weight (Volesky *et al.*, 1983). Cadmium resistance in *Staphylococcus aureus* is, for example, plasmid-bound, due to an increased Cd-efflux system [2 Cd<sup>2+</sup>/2H<sup>+</sup> antiporter (see Foster, 1983)]. Arsenate resistance is also plasmid-bound and linked to arsenite and antimony resistance (Foster, 1983). In *Pseudomonas putida*, cadmium resistance is due to synthesis of three novel proteins rich in cysteine [MW 4000–7000 (see Higham *et al.*, 1985)].

Information on the physiological effects of most of these potentially toxic regolith components on microorganisms is scanty. Although some of the elements listed in Table 4 are probably needed as "trace elements," the concentration ranges offered to the bacteria are extremely critical. Therefore, experiments will have to be conducted in which a large number of bacterial test strains are exposed to various lunar soil samples under simulated lunar reactor conditions.

## SOME FINAL THOUGHTS

At the present time, it does not seem to be possible to send experimental cultures of bacteria to the Moon to study their longevity and activities under lunar conditions. However, the "Biostack" experiments of the Apollo 16 and 17 missions have shown that living microorganisms can indeed be carried safely to the lunar surface and back (Taylor, 1974). *Bacillus subtilis* spores embedded in polyvinyl alcohol sheets were subjected to high-energy, multicharged ("HZE") particles, as indicated by tracks registered in special dosimeters. After this treatment, the *Bacillus subtilis* spores germinated, grew, and elongated just as well as the untreated ground controls did.

The quantitative aspects of the hydrogen-harvesting processes are difficult to assess. We assume an average bacterial cell composition as described in Table 5. We can further assume for the sake of calculation that respiration or other processes would not result

Table 5. Composition of an Average, Growing Bacterium

Water	80 %	
Dry matter	20 %	
carbon		45–55 %
oxygen		20 %
nitrogen		10–15 %
hydrogen		10 %
phosphorus		2–6 %
other elements		5 %

From Schlegel, 1984.

in loss of cell substance during the synthesis of 1 g of microbial biomass. Living bacterial substance should then be composed of water (800 mg/g) and dry matter (200 mg/g). This dry biomass would then consist of approximately 50% (or 100 mg) carbon, 20% (or 40 mg) oxygen, 10% (or 20 mg) nitrogen, 10% (or 20 mg) hydrogen, 5% (or 10 mg) phosphorus, and 5% (or 10 mg) of other elements.

To extract 20 mg hydrogen from an average lunar soil (size fraction  $< 20 \mu\text{m}$ , containing 100 ppm hydrogen), we would need 200 g regolith fines. Since energy is indeed required to construct a living organism, one might have to spend perhaps 1–2 kg of lunar dust for the production of 1 g of living bacteria. With respect to carbon, and assuming that 5–10 g of soil contain 1 mg C, the construction of 1 g living bacteria would require at least 0.5–1.0 kg of soil. The availability of carbon for the bacteria has not been considered in the calculation, but elemental carbon has not been known to be utilized by bacteria. For the nitrogen, 200 g of soil would probably be sufficient. In total, 3–5 kg of lunar soil would allow the construction of 1 g living bacteria—always assuming that the elements can be offered in a “palatable” form.

The availability of versatile bacteria with many different metabolic possibilities for the utilization of lunar regolith hydrogen has been demonstrated. Next should be an assessment of actual yields of these reactions under (simulated) lunar conditions. Combinations of different organisms to form active consortia or syntrophic mixtures would have to be tested for optimal metabolic activity and longevity. Genetic engineering might be necessary to “train” the selected bacteria for tolerances to lunar stress conditions. Thus, artificial microcosms (ecosystems) will have to be created, and their homeostasis (ability to return to original activity after a perturbation) will have to be optimized, possibly by combining a large number of different microorganisms to perform a given reaction. This latter activity represents a challenge to present-day microbiologists who plan future lunar colonies.

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# HYDROGEN AND WATER DESORPTION ON THE MOON: APPROXIMATE, ON-LINE SIMULATIONS

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To help assess possible water reserves on the Moon derived from the solar wind (which could supply a lunar base) silicon, sapphire, and oligoclase (feldspar) have been irradiated with a beam of 2.5 keV/amu deuterium ions to simulate solar wind proton bombardment of lunar materials. For silicon and sapphire the areal density of deuterium  $D(\phi)$  (atoms/cm<sup>2</sup>) increases with the incident ion fluence  $\phi$  until a critical re-emission fluence is reached ( $\phi_{re} \approx 3 \times 10^{17}$  D/cm<sup>2</sup>). At this point deuterium begins to be lost from both targets until  $D(\phi)$  reaches a saturation value  $D_s \approx 3 \times 10^{17}$  D/cm<sup>2</sup> at  $\phi_s \approx 10^{18}$  D/cm<sup>2</sup>. Oligoclase begins losing deuterium when the beam is turned on, and  $D(\phi)$  never reached a saturation value up to the limit of our experiment ( $\phi \approx 3 \times 10^{18}$  D/cm<sup>2</sup>). There was also a small yield of  $\sim 10^{-4}$  D<sub>2</sub>O<sup>+</sup>/D<sub>incident</sub> during the implantation of oligoclase. The similar saturation values for the dissimilar materials silicon and sapphire and the lack of a saturation value for oligoclase are unexplainable by models previously used to describe the build-up of solar wind species in lunar samples. Also, the thermal release patterns for our targets have three generally lower temperature peaks when compared to those of returned lunar soils. These differences may be the result of imperfect simulation conditions or the hydration of the returned samples.

## INTRODUCTION

It has been argued that solar wind (SW) hydrogen implanted into lunar dust grains could result either in the storage of hydrogen by OH bonds in oxygen-rich grains (Zeller *et al.*, 1966) or in the release of small molecules such as H<sub>2</sub> and H<sub>2</sub>O during pyrolysis of lunar soil samples (Gibson and Moore, 1972; Desmarais *et al.*, 1974; Bustin *et al.*, 1984). The latter authors found that  $\sim 100$  ppm water is released from lunar soil samples. At this level of abundance, water will have to be found at the polar regions of the Moon (Watson *et al.*, 1961; Arnold, 1979) or hydrogen will have to be brought from Earth for maintaining life at a lunar base. We believe that it is useful to study the uptake and release of SW hydrogen in lunar analog materials to assess whether there are components of the lunar soil containing a much higher concentration of SW hydrogen that may be economically extractable and also to provide data to better estimate the SW contribution to possible lunar ice reservoirs.

The first step in this assessment is to question the validity of the basic models that have been used over the last 15 years for describing the accumulation and release of



SW-related species at lunar conditions. These models have been thoroughly reviewed by Pillinger (1979). In the most frequently quoted model, it has been assumed that the surface concentration  $C_s$  of an implanted species reflects the value of the implant fluence required to sputter away a thickness of target material corresponding to the projected range of the incident ions, that is,  $C_s \propto Y_s^{-1}$ , where  $Y_s$  is the sputtering rate of the incident ions (Carter and Colligon, 1968; Carter *et al.*, 1972). Another model requires that the areal density  $D_s$  of the implanted species be somewhat scaled by the value of the amorphizing fluence  $\phi_a$ , which forms an amorphous layer on most lunar silicate minerals (Bibring *et al.*, 1974b). In this paper we present recent results concerning the validity of these models, which were obtained by using a reaction chamber developed in fusion reactor research. Our results show that the models for accumulation and release of SW species do not appear to be totally valid and suggest that mature lunar surface soils may act as efficient converters for transforming the flux of energetic SW protons into a flux of low energy hydrogen and water molecules continuously injected into the lunar atmosphere.

## EXPERIMENTAL DETAILS

### Reaction Chamber

The reaction chamber used in this work was developed a few years ago for investigating the retention in the near-surface range, the diffusion into the bulk material, and the release of hydrogen isotopes from metals (Scherzer *et al.*, 1983). This chamber is equipped with a quadrupole mass spectrometer on-line with both a 30 kV accelerator for ion implantation and a 2.5 MV Van de Graaff accelerator for analysis. The mass spectrometer can monitor the simultaneous release of four distinct masses in the mass range 1–100 during ion implantation and during thermal desorption runs when the target can be heated up to  $\sim 900^\circ$  C. To minimize mass interferences associated with terrestrial contaminants, which yield strong backgrounds at masses 1, 2, 3, 18, and 20, we implanted with  $^2\text{D}$  instead of  $^1\text{H}$  at an energy of 5 keV/atom. We simulated the effects of He-damage, which create H trapping sites, with  $^3\text{He}$  pre-implants. We monitored the release of  $\text{D}_2$  and  $\text{D}_2\text{O}$  molecules. The residual concentration as well as the depth concentration profile of deuterium in the targets were measured using the  $\text{D}(^3\text{He}, \alpha)\text{H}$  nuclear reaction with a  $\sim 1 \text{ mm}^2$  beam of 750 keV  $^3\text{He}$ . It is worth noting for our subsequent discussion of water generation at lunar conditions that the vacuum and the residual  $\text{H}_2\text{O}$  pressure in the chamber were  $\sim 10^{-8}$  and  $\sim 10^{-10}$  mbar, respectively.

### Choice of Targets

Except for the work of Arnold and Doyle (1982) on fused silica, only materials relevant to fusion reactor technology, such as metals (Möller, 1983) and some “poor” conductors such as graphite (Sone and McCracken, 1982), TiC (Doyle *et al.*, 1981), and  $\text{TiB}_2$  (Doyle and Vook, 1979), have been investigated using sophisticated “on-line” techniques. For our investigation we have chosen terrestrial analogs of lunar materials and silicon and sapphire. In this paper we report our early results for silicon, sapphire, and oligoclase.

Most of the lunar surface is covered with amorphous and crystalline silicates that belong to the specific family of insulators known as nuclear track detectors. These solids register tiny regions of radiation damage along the path of both high ( $E \approx 1$  MeV/amu) and low energy ( $E \approx 1$  keV/amu) ions. At a particular critical implant dose  $\phi_a$ , they develop an amorphous coating of radiation damage showing a drastically increased chemical reactivity. The hydration ability of this coating can be several orders of magnitude greater than that of undamaged material (Dran *et al.*, 1984). Because planetary scientists have assumed that radiation damage or sputtering rates play a dominant role in the accumulation of SW species in the lunar regolith, we have used targets of silicon and sapphire, which, as well as the metals and poor conductors, are different from the solids classed as nuclear track detectors. Their stability against radiation damage spans  $>1000$  fold range (Naguib and Kelly, 1975), and their sputtering rates span  $>10$  fold range compared to those of the lunar nuclear track detectors (Carter and Colligon, 1968).

## PRELIMINARY RESULTS AND DISCUSSION

### Build-up of Saturation Concentrations: Results

During the 2.5 keV/amu deuterium implantation at room temperature, we continuously monitored the variation of the areal density  $D(\phi)$  (atom/cm<sup>2</sup>) of deuterium retained in the targets as a function of the incident ion fluence  $\phi$ . A simple and similar trend is observed for both silicon and sapphire. At first the areal density of D in the target linearly increases with  $\phi$ , with no D<sub>2</sub> re-emission being detected with the on-line mass spectrometer. At a critical re-emission fluence  $\phi_{re} \approx 3 \times 10^{17}$  D/cm<sup>2</sup>, deuterium begins to be lost from the targets. Finally  $D(\phi)$  reaches a saturation value  $D_s \approx 3 \times 10^{17}$  D/cm<sup>2</sup> at  $\phi_s \approx 10^{18}$  D/cm<sup>2</sup>. The values of  $D_s$  and  $\phi_s$  noted for silicon and sapphire are similar. It is remarkable that this behavior corresponds to that of metals at liquid nitrogen temperature (Möller, 1983) and of fused silica at room temperature (Arnold and Doyle, 1982) with approximately the same saturation fluence  $\phi_s \approx 10^{18}$  D/cm<sup>2</sup>. On the other hand, oligoclase drastically departs from this simple behavior and loses deuterium at a low fluence ( $\sim 10^{15}$  D/cm<sup>2</sup>), that is, when the beam is turned on. The areal density  $D(\phi)$  continuously increases but never reaches a saturation value up to the limit of our experiment at  $\phi \approx 3 \times 10^{18}$  D/cm<sup>2</sup>. The continuous loss of deuterium from oligoclase is possibly related to the "percolation" of deuterium through radiation damage islands produced by ion implantation. This mechanism has been proposed to explain the greatly enhanced hydration ability of ion-implanted silicates when exposed to water or water vapor (Borg *et al.*, 1982). They found that the increased hydration ability begins with extremely low fluences of implanted hydrogen ( $\sim 10^{-2} \phi_a$ ).

### Build-up of Saturation Concentrations: Inadequacy of Previous Models

According to proposed models, the saturation concentration  $C_s$  should vary inversely with the sputtering rate  $Y_s$  (Carter and Colligon, 1968; Carter *et al.*, 1972), or the areal density  $D_s$  of the implanted species should vary with the amorphizing fluence  $\phi_a$  (Bibring *et al.*, 1974b). But the targets used in our experiments clearly have similar values of

$C_s$  and  $\phi_a$  despite their drastic differences in sputtering rates and stability to radiation damage. Consequently, models claiming a simple relationship between  $C_s$  and  $Y_s$  or  $D_s$  and  $\phi_a$  require revision. Our depth profile measurements indicate that the bulk concentration of hydrogen is limited to a fixed value that depends on the material and the re-emission of excess atoms. In this work we find that this maximum concentration is  $\sim 0.3$  D atom/matrix atom.

### **Thermal Desorption of Hydrogen Isotopes in Terrestrial Targets**

The thermal release patterns of silicon, sapphire, and oligoclase were obtained by continuously monitoring the release of  $D_2^+$  ions with the mass spectrometer while the sample was being heated (Fig. 1). These patterns show at least three distinct peaks and are more complex than the single peak structure observed for metals and fused silica. This multi-peak structure, which signifies complex D-traps in the material, is unique for each solid as opposed to the approximately constant values of  $\phi_s$ . Furthermore, heating the material well below melting temperature totally extracts the trapped deuterium. After the highest temperature release peaks, observed at  $\sim 690^\circ\text{C}$ ,  $\sim 830^\circ\text{C}$ , and  $\sim 430^\circ\text{C}$  for silicon, sapphire, and oligoclase, respectively, no deuterium can be detected in the samples with the  $^3\text{He}$  analysis beam. This type of behavior has also been observed for metals (Möller, 1983), graphite (Sone and McCracken, 1982), TiC (Doyle *et al.*, 1981), and  $\text{TiB}_2$  (Doyle and Vook, 1979) implanted at low temperatures and fused silica implanted at room temperature (Arnold and Doyle, 1982).

### **Thermal Desorption of Hydrogen Isotopes in Lunar Soil Samples**

The thermal release patterns observed in our experiments are markedly different than those observed for lunar soil samples (Fig. 1). The thermal release pattern shows two distinct peaks at  $\sim 600^\circ\text{C}$  and  $\sim 1200^\circ\text{C}$  (*i.e.*, at melting temperature) for most solar wind related species (including hydrogen) (Gibson and Moore, 1972; Desmarais *et al.*, 1974; Bustin *et al.*, 1984). Clearly it is necessary to investigate whether integrated temperature release peaks observed in our preliminary work can at least model the low temperature peak found in lunar soils and also whether our low temperature peaks can be shifted to higher temperatures by additional effects. Some of these effects include lunar conditions that have not been simulated such as implantation at the effective day temperature of the Moon ( $\sim 130^\circ\text{C}$ ) (Taylor, 1975), predamage associated with other SW implanted species, especially helium, the chemisorption of hydrogen on SW He bubbles tightly bound to structural defects, and the multiple implants of H, N, C, and other SW ions. An earlier work indicates that the pyrolysis of feldspar targets subjected to multiple implants with  $^2\text{D}$ ,  $^{15}\text{N}$ , and  $^{13}\text{C}$  ions better reproduces the two peak release pattern observed for lunar soils (Bibring *et al.*, 1974a). Another consideration is the increased hydration ability of hydrogen implanted silicates (Dran *et al.*, 1984). The best preserved lunar soil samples have been kept in "dry" nitrogen that still contains  $\sim 200$  ppm residual water. Because Dran *et al.* (1984) found that hydrogen implanted silicates have increased hydration abilities, this water may quickly permeate the entire depth of the amorphous coatings formed on lunar silicates and mix with the SW trapped species (Leich *et al.*, 1974). The

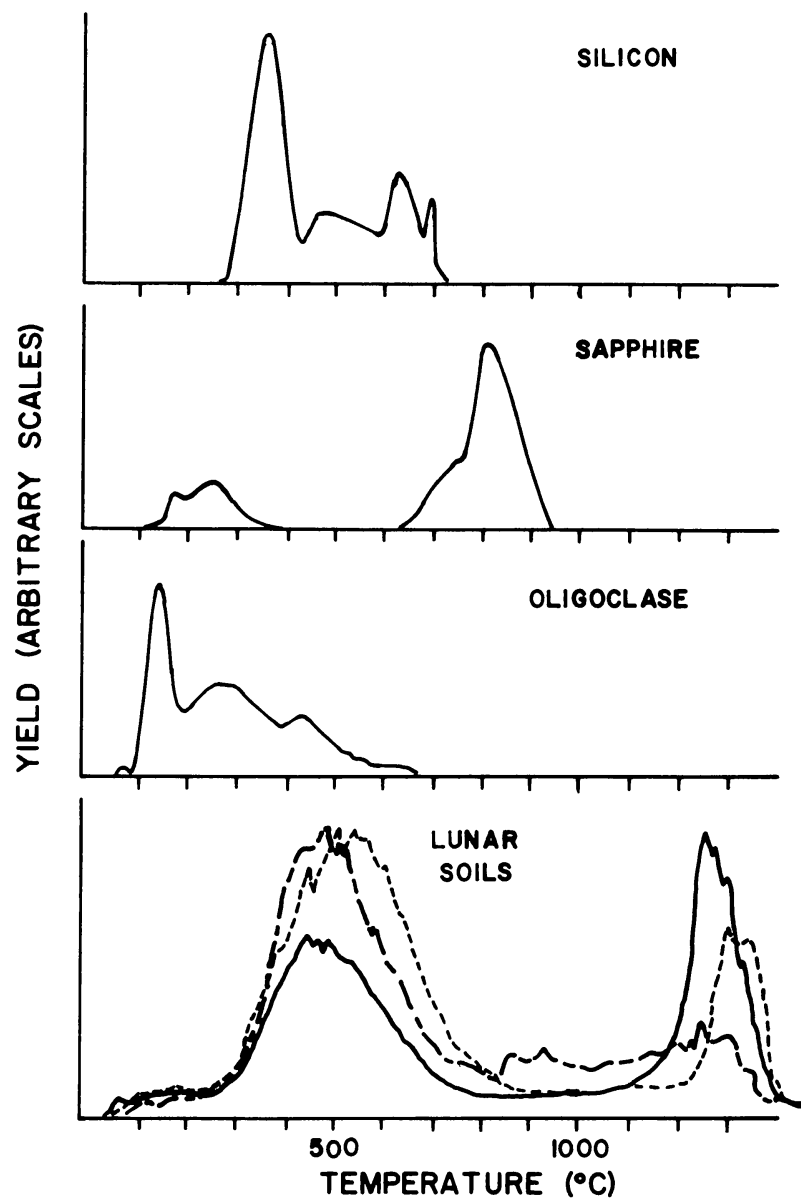


Figure. 1. Thermal desorption of implanted  $D_2$  from silicon, sapphire, and oligoclase and of solar wind  $H_2$  from lunar soils [solid line = 14163,178, dotted line = 15021,21, dot-dash line = 15601,31; from Gibson and Moore (1972)].

resulting hydrated layer may form new families of high temperature traps that would behave differently than those in truly pristine lunar samples and our targets. It has also been suggested that the high temperature peak may result from SW hydrogen redistributing deeply into agglutinates during their formation (D. S. McKay, personal communication, 1984). In previous thermal release experiments on lunar soil samples it has been assumed that terrestrial water would be weakly adsorbed on the grain surfaces and would result in the lowest temperature release peak, which would be easily distinguishable from the high temperature release peaks expected for SW hydrogen. Because hydration ability has been found to be increased by several orders of magnitude by radiation damage (Dran *et al.*, 1984), we believe that there is reason for questioning this assumption. This would imply that the value of  $\sim 100$  ppm SW hydrogen published for lunar soil samples needs to be reassessed.

### **The Lunar Surface: A Solid State Converter for the Continuous Generation of $H_2$ and $H_2O$ Molecules in the Lunar Atmosphere**

We have reported on the re-emission of the  $D_2$  molecules from all targets during either ion implantation at room temperature or during thermal desorption runs at  $T > 150^\circ C$ . We have also detected a small yield of  $\sim 10^{-4} D_2O^+/D_{\text{incident}}$  during the implantation of oligoclase starting at a low implantation fluence. To be sure that we were not measuring the "memory" of the chamber walls to previous experiments, we implanted  $^3He$  in a similar target pre-implanted with a high dose of D. This target released a much lower yield of  $D_2O$  from the D implanted, oxygen-rich targets. If lunar silicates behave like oligoclase, the re-emission of  $H_2$  and  $H_2O$  would be initiated at low SW ion fluences. Consequently, during the lunar day, areas of mature regolith on the lunar surface, which are estimated to be about 1/3 of the total surface (Langevin, 1978), may act as efficient "converters" for transforming the flux of SW protons ( $\sim 3 \times 10^8 \text{ H.cm}^{-2}.\text{s}^{-1}$ ) into an outward flux of low energy  $H_2$  and  $H_2O$  molecules. This continuous outgassing of the lunar regolith may contribute significant amounts of water to permanently shadowed regions near the lunar poles (Watson *et al.*, 1961; Arnold, 1979) and may initiate a weak hydration of the lunar regolith materials. Such a water cycle may have implications for other astrophysical sites as well.

*Acknowledgment*      This work has been supported in part by NASA grant NSG 9043.

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# AN ANALYSIS OF ALTERNATE HYDROGEN SOURCES FOR LUNAR MANUFACTURE

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Earth's relatively large mass places a high energy cost on the use of terrestrial materials in space. The availability of minerals on the Moon at one-sixth Earth gravity, coupled with ample solar energy, ensures that some materials required for permanent lunar bases and space exploration will be manufactured there. Terrestrial hydrogen must be imported in some form to supply the critical need for water and oxygen for life support until lunar vacuum pyrolysis is established and thereafter, as well, if the supply of lunar hydrogen is inadequate. The importation of liquid hydrogen is generally considered, but liquid methane and liquid ammonia should also be evaluated. A weight penalty is associated with importing hydrogen as methane or ammonia; however, higher boiling points reduce boil-off during transport and storage, offsetting this penalty. In addition, methane and ammonia can be chemically converted into other compounds by industrial processes for which effective catalysts have been developed, and they can be used for reduction of ilmenite.

## INTRODUCTION

A limitation to the exploration and use of space for scientific and industrial enterprise is the high cost of energy needed to overcome the Earth's gravitational field in order to lift terrestrial materials into space. The reusable space shuttle reduces the capital cost of ferrying such materials to near-Earth orbit but has little impact on the energy cost. The material requirements of servicing satellites in geosynchronous orbit and of exploring and colonizing the Moon and nearby planets will be even more energy-expensive if all that material must be of terrestrial origin. One cost-reducing alternative is to use materials already available in near-Earth space, that is, on the Moon and on asteroids with Earth-approaching orbits (Steurer, 1982).

The availability of minerals on the Moon, coupled with ample solar energy, ensures that some materials required for permanent lunar bases will be manufactured there. The lower energy cost of material in space, resulting from the one-sixth lower gravity of the Moon compared with the Earth, assures the use of lunar materials for space exploration as well.

Lunar material has been sampled by astronauts from the U.S. Apollo missions and by automatic remote samplers of the Soviet Union. From extensive chemical analysis of lunar rocks brought back to Earth, considerable information is available on the chemical

Table 1. Lunar Rock Composition

Major elements, > 1%	Minor elements, > 0.1%	Trace elements
O	Na	C
Si	Cr	N
Al	K	F
Fe	Mn	Ni
Ca	P	Ba
Mg	S	Sr
Ti		Zr
		H
		He
		Cl
		Co
		all others

composition of lunar surface rocks and soil of both mare and highland areas (Arnold and Duke, 1977; Criswell, 1983a).

Table 1 lists the elements found in lunar material in order of abundance (Criswell, 1983b; Phinney *et al.*, 1977). Oxygen is the most abundant, followed by silicon, aluminum, and iron. Carbon, nitrogen, and hydrogen are found only in trace amounts in the upper layers of soil. Their presence is attributed to the deposition of ions from the solar wind and possibly from meteorites. All of the other elements found on Earth are also present in varying but limited amounts.

The most desirable materials for early lunar manufacture are those required for life support, shelter, construction, and rocket propulsion. Using available lunar elements to meet these needs suggests that the first materials to be manufactured on the Moon will be water, oxygen, iron, titanium, silicon, aluminum, and ceramic heat and radiation shields (Phinney *et al.*, 1977). Water and oxygen are essential for life support. In addition, oxygen is an important component for chemical rocket propulsion. It accounts for the major mass needed for space transportation in the vicinity of Earth. Looking ahead to a permanent lunar colony suggests that carbon and nitrogen compounds will be needed for plant and animal life support.

To make water and important carbon and nitrogen compounds requires hydrogen. In addition, hydrogen is an effective reductant for lunar minerals for production of oxygen and metals. Therefore, this analysis of alternate hydrogen sources for lunar manufacture should be of value in planning for future missions for manned exploration and settlement of the Moon.

## LUNAR PRODUCTION OPTIONS

Water is needed to support life. Only traces of water have been found on the Moon (Criswell, 1983a). Free water vapor is precluded by the near-vacuum of the lunar atmosphere with a pressure of less than  $10^{-12}$  torr in the daytime and less than  $10^{-14}$  at night (Criswell,



1983a). Soil at the lunar surface holds between 50 and 100 ppm of water originating from reaction of solar wind protons with lunar oxide minerals (Criswell, 1983a). The water can be freed by vacuum pyrolysis at 200°–600°C. There is less than 10 ppm of bound water in the lunar rocks, which requires temperatures above 1000°C for release (Criswell, 1983a).

Vacuum pyrolysis yields other volatile components besides water (Gibson and Hubbard, 1972). Any free hydrogen in the volatile components can be oxidized to water by reduction of suitable lunar mineral oxides such as ilmenite. However, vacuum pyrolysis would require mining and handling more than four metric tons of lunar soil for each gallon of water produced.

A detailed analysis has been made by Arnold (1979) of the possibility that water may accumulate in cold traps at the lunar poles. Areas in perpetual darkness in polar craters and rock crevices are estimated never to exceed about 100 K. Some of the water vapor released on sunlit lunar surfaces at above 400 K, because of the long mean free path for gases in the high vacuum lunar atmosphere, would quickly diffuse to the traps and be held there for many millenia. Confirmation of the speculation on trapped ice awaits exploration of the lunar poles.

Obviously, water used on the Moon, whether found, produced, or imported, would be carefully conserved, purified, and recycled. During the early stages of lunar base development and if the lunar sources prove inadequate, water will have to be imported. To have water on the Moon, only the hydrogen portion need be imported since oxygen is available there. Alternate options for hydrogen import to the Moon will be evaluated.

Because both carbon and nitrogen are critical to animal and plant life, adequate supplies must be available to maintain permanent lunar colonies. The solar wind deposits both in lunar surface soil. Carbon has been found in lunar rocks at 20–200 ppm and freed by vacuum pyrolysis at temperatures up to 600°C as carbon dioxide and methane and above 1000°C as carbon dioxide and carbon monoxide (Criswell, 1983a). Similarly, 50–150 ppm of nitrogen has been found and isolated as molecular nitrogen by vacuum pyrolysis at above 1000°C (Criswell, 1983a). If vacuum pyrolysis proves inadequate, terrestrial C and N will have to be imported. The simplest form for their import is combined with hydrogen as methane and ammonia.

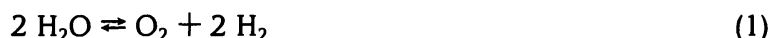
There is ample oxygen on the Moon in the form of oxidic minerals (Arnold and Duke, 1977; Criswell, 1983a). Because oxygen is so critical for life support and is a major component by weight for rocket propulsion, its production has received considerable attention. Oxygen probably will be the first lunar resource to be exploited for space exploration.

Oxygen can be isolated by direct pyrolytic or electrochemical treatment of lunar silicates (Criswell, 1978; Jarrett *et al.*, 1980). Other methods, which require importing reagents from Earth, include carbo-chlorination and HF, acid, or alkaline decomposition of silicates followed by electrochemical separation of components (Criswell, 1978; Jarrett *et al.*, 1980). More desirable methods are based on the reduction of lunar minerals, such as ilmenite, followed by electrolysis of the water produced to yield oxygen along with hydrogen, which can be recycled to the reduction process (Williams, 1983).

Hydrogen compounds are required for reduction of ilmenite. Prior to lunar application of vacuum pyrolysis, or if the lunar hydrogen supply is inadequate, some hydrogen will have to be imported from the Earth. The importation of liquid hydrogen is generally favored, but alternate hydrogen sources such as liquid methane and liquid ammonia should also be evaluated.

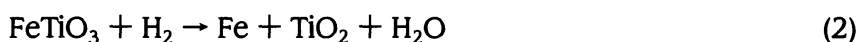
## LUNAR HYDROGEN OPTIONS

A source of hydrogen will be needed on the Moon by explorers or colonizers to ensure an ample supply of water. With water available, a supply of oxygen can also be ensured. The decomposition of water into its elements, hydrogen and oxygen, is shown as:



The forward reaction can readily be carried out electrochemically to produce oxygen along with hydrogen for recycle. Membranes, catalysts, electrodes, and other equipment for electrolysis are commercially available because the power generating industry produces hydrogen for use as a gaseous lubricant for high tolerance electric generator bearings in this way (Nuttall, 1980).

The reverse reaction to produce water can be carried out by reacting hydrogen with oxygen in a suitable burner. However, it is better to use the hydrogen for direct reduction of lunar oxide minerals to produce water and metal from the mineral. This reaction is illustrated below in (2) for the reduction of ilmenite, an iron titanate mineral found in ample supply in the lunar mare regions (Arnold and Duke, 1977).



Hydrogen reduction of ilmenite has been studied (Williams, 1983; El-Guindy and Davenport, 1970). High temperature is required to drive the reaction to the right. Further development is likely to yield catalysts that accelerate the reaction at lower temperatures. The products of this reaction, all useful on the Moon, are metallic iron, titania, which has a high albedo, and water for direct use or for electrolysis to oxygen and hydrogen. The hydrogen can then be recycled for further ilmenite reduction.

Clearly, lunar hydrogen and water will require considerable mining and material handling for vacuum pyrolysis of lunar surface particles to yield the small percentage they contain. Also, until further exploration is undertaken, the existence of trapped water at the lunar poles must be considered speculative. If these lunar sources prove to be inadequate, or too costly to develop, hydrogen will have to be imported from Earth.

The most convenient form for shipment of terrestrial hydrogen to the Moon is as liquified gases such as molecular hydrogen, methane, and ammonia. Relevant properties of these liquids are in Table 2.

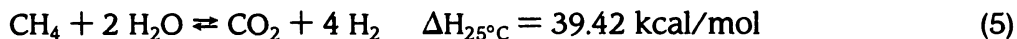
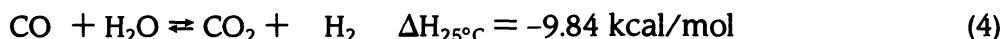
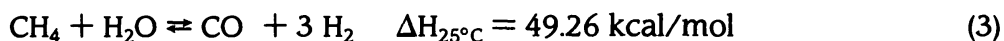
Hydrogen can be obtained from methane with carbon monoxide and carbon dioxide as by-products. The petrochemical industry applies the water/gas shift reactions to reform

Table 2. Hydrogen Imports

	Hydrogen	Methane	Ammonia
Molecular weight	2	16	17
Hydrogen % by weight	100	25	17.6
Boiling point, °C	-252.8	-161.5	-33.35
Liquid density at bp, g/ml	0.070	0.424	0.68
Hydrogen weight at bp, g/ml	0.070	0.106	0.120
Heat of vaporization at bp, J/g	452.2	577.8	1369

methane for production of hydrogen, carbon monoxide, and carbon dioxide from natural gas (Richardson, 1973; Czuppon and Buividas, 1980). Most of the industrial hydrogen used for hydrogenation and to make ammonia is produced by this chemistry.

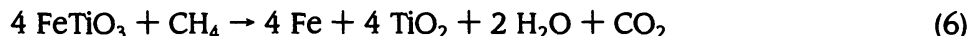
The conditions of temperature, pressure, concentration, contact time, and catalyst variations to allow industrial interconversion of these compounds are well known (Bridger and Chinchin, 1970). This chemistry is illustrated in (3), (4), and (5).



Chemical engineers have developed proven industrial processes and plant designs for carrying out these interconversions (Moe, 1962). Adapting this terrestrial chemical technology to lunar conditions will require extensive further development.

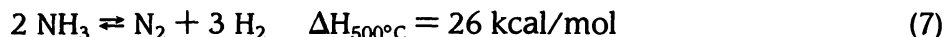
Chang (1959) has shown that ilmenite can be reduced to iron in 80% yield in one hour at 1100°C with carbon monoxide, hydrogen, or their mixtures. Russian scientists, because of the large deposits of ilmenite and natural gas found in Siberia, have studied the reduction of ilmenite with methane (Reznichenko *et al.*, 1983). They obtained 85–90% reduction of ilmenite as 0.25–0.5 mm particles with natural gas in a fluidized bed at 1000°–1030°C in 5–7 minutes.

Although, methane may be undergoing the reforming reactions of (3), (4), and (5), and the ilmenite reduction is actually effected by carbon monoxide and/or hydrogen, the overall reduction of lunar oxidic minerals such as ilmenite by methane may be summarized by (6):



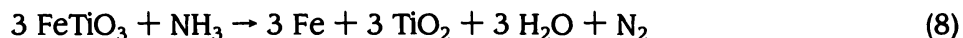
Reaction (6) does yield products useful on the Moon, namely iron, titania, water, and carbon dioxide. Further extensive development will be required to adapt the Russian process to lunar conditions.

Hydrogen can also be readily freed from ammonia by reversal of the ammonia synthesis reaction, (7).



Synthesis is favored by low temperature and high pressure and decomposition by high temperature and low pressure (Bridger and Snowdon, 1970). Iron catalyzes both the ammonia synthesis and reverse reactions (Czuppon and Buividas, 1980). The small amount of particulate metallic iron, which can be separated magnetically from the lunar surface soil, may well be an effective catalyst (Steurer, 1982). The nitrogen produced can be used to dilute oxygen for breathing or oxidized to nitric oxides electrochemically. The product of these nitric oxides and additional ammonia is ammonium nitrate, an effective fertilizer for plants and an explosive.

However, it may prove possible to find the proper conditions of temperature and pressure to control the ammonia-hydrogen equilibrium in order to reduce ilmenite directly with ammonia as illustrated in (8).



Otherwise, the hydrogen from the ammonia decomposition can reduce the ilmenite directly according to (2) with the nitrogen only acting as a diluent. Again, development of the necessary reaction conditions and engineering for the lunar environment will require further study.

## CRITERIA FOR CHOICE OF HYDROGEN IMPORT

A weight penalty is associated with importing hydrogen as methane or ammonia compared with molecular hydrogen (see Table 2). For every ton of hydrogen imported as methane, three tons of carbon must also be imported; for every ton of hydrogen imported as ammonia, 4.7 tons of nitrogen must be imported. If the carbon or nitrogen would have to be imported anyway, bringing hydrogen along as methane or ammonia is reasonable, since other factors offset part of the weight penalty. For example, based on liquid density at the boiling point, liquid hydrogen requires a vessel 9.7 times larger than that for an equal weight of liquid ammonia, or 6 times larger than one for liquid methane.

Based on hydrogen content alone, the vessel to transport one ton of hydrogen as liquid hydrogen would be 1.5 times larger than one to transport a ton of hydrogen as liquid methane and 1.7 times larger than one to transport a ton of hydrogen as liquid ammonia. Thus, for the same hydrogen payload, the additional weight of the carbon or nitrogen is partly offset by the lighter weight of a smaller vessel with thinner walls.

In addition, the higher boiling points of methane and ammonia compared with hydrogen helps offset the weight penalty through reduced boil-off during transport and storage and simpler containment, insulation, and refrigeration systems. A simpler container and a lower insulation requirement translates into less container weight for equal boil-off.

This effect is enhanced by the three-fold higher heat of vaporization in joules per gram of ammonia at its boiling point compared with hydrogen at its boiling point. The heat of vaporization of methane is 1.3 times that for hydrogen.

The temperature on the surface of the Moon varies from 400 K during the lunar day to 100 K during the lunar night (Criswell, 1983a; Williams and Jadwick, 1980). At a depth of 150 cm below the surface, the temperature is estimated to be nearly constant at about 253 K (Langseth, 1976). The boiling point of liquid ammonia is just 13 K lower, so that a buried liquid ammonia pressure vessel with no boil-off can be considered. The boiling point of liquid methane is 141 K and of liquid hydrogen is 233 K lower than this subsurface temperature. The larger the differential, the faster the boil-off, or the more insulation required.

## CONCLUSION

For sustained lunar colonization, carbon and nitrogen will be needed for plant and animal growth and for production of useful synthetic organic materials. Lunar minerals contain little carbon and nitrogen. Since they may have to be imported, a useful way to import them could be as methane and ammonia. Methane can be converted into carbon dioxide, carbon monoxide, and hydrogen by well known industrial processes for which effective catalysts have been developed, and it can also be used directly for reduction of ilmenite. Ammonia can be converted to nitrogen and hydrogen. Carbon dioxide fed directly to plants along with ammonia and its by-products form the essential chemicals required for sustained plant life in a self-sufficient lunar habitat.

The well developed industrial chemical processes for utilizing these gases on Earth need to be adapted for lunar operation. For example, carbon monoxide and hydrogen can produce a wide array of hydrocarbons via such synthetic gasoline processes as Fischer-Tropsch (Dry, 1976; Vannice, 1976). Thereby, broad vistas of well known organic chemical processes needed for life on the Moon are opened.

Based on known chemistry, the complex task of engineering a process chemical industry for automated operation in the lunar environment is essential to colonization of the Moon.

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