

CARBOTHERMAL REDUCTION OF LUNAR MATERIALS FOR OXYGEN
PRODUCTION ON THE MOON
REDUCTION OF LUNAR SIMULANTS WITH CARBON

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The utilization of extraterrestrial resources will become a key element in space exploration and colonization of the Moon and Mars in the 21st century. Indeed, the development and operation of *in-situ* manufacturing plants are required to enable the establishment of permanent lunar and Martian bases. Oxygen manufacture for life support and propulsion will be the most important manufacturing process for the first of these plants. The Carbothermal Reduction Process for the manufacture of oxygen from lunar materials has three essential steps: the reduction of ferrous oxide and metallic silicates with methane to form carbon monoxide and hydrogen; the reduction of carbon monoxide with hydrogen to form methane and water; and the electrolysis of water to form oxygen and hydrogen. This closed cyclic process does not depend upon the presence of water or water precursors in the lunar materials. It produces oxygen from silicates regardless of their precise composition and fine structure. In accord with the Statement of Work of Contract NAS 9-19080, Carbothermal Reduction of Lunar Materials for Oxygen Production on the Moon, ORBITEC has placed emphasis on the following issues to gain a better understanding of the Carbothermal Reduction Reaction of lunar regolith and to develop a low-risk, light-weight design for a lunar lander experiment: (1) reduction of lunar simulants with carbon (or equivalent); (2) determination of conditions, particularly temperatures, required for initial and complete reduction of lunar simulants by the carbon-containing reducing agents; (3) identification of the products formed, gases and solids; (4) determination of solid product properties; (5) determination of reaction rates and mechanisms; (6) selection and demonstration of container materials; and (7) selection and demonstration of heating methods. The most important of these issues were: (1) reduction of lunar simulants, i.e., JSC-1, MLS-1A, Ilmenite, and Gruenerite, with carbon, i.e., graphite; (2) identification of the products formed, i.e., carbon monoxide, metals, e.g., iron and silicon, and slags, e.g., complex silicates; (3) selection and demonstration of container materials, e.g., stabilized zirconia and yttria; and (4) selection and demonstration of heating methods. The results of this empirical research are reported in this paper.

The following activities were conducted: (1) experiment requirements definition; (2) lunar simulant acquisition; (3) small furnace apparatus design; (4) apparatus fabrication, assembly and checkout; (5) experiment execution; and (6) reduction product analysis and identification. This paper will focus on (1) experiment execution and (2) reaction product analysis and identification.

The main components of the test apparatus are the high-temperature furnace (HTF), the quadrupole mass spectrometer (QMS), and the electronic mass-flow control system. The HTF is a vacuum-tight, water-cooled, resistance furnace with graphite heating elements capable of reaching a maximum temperature of 2200 C (3992 F). The temperature was controlled using a state-of-the-art, programmable temperature controller. The temperature control thermocouple (boron/graphite design) is recommended for use up to 1800 C (3272 F). The temperature can be controlled to remain constant at a given level to an estimated average of ± 1 C (1.8 F). The HTF was mounted so it can be rotated from horizontal to vertical in any required orientation. It has a working space of 11.43 cm (4.5 in.) dia by 15.24 cm (6 in.) high (vertical position). It was maintained in the horizontal position in all of the experiments to provide a surface which could accommodate as many as six crucibles. The QMS used for product gas analyses was placed next to the HTF. Two electronic mass-flow controllers were used to control and measure flow, one each for inlet and exit gases. All of the gas lines were made of 6.35 mm (0.25 in.) OD stainless steel tubing. The crucibles used were all of the same capacity and dimensions, i.e., 50 ml capacity, 35 mm (1.38 in.) dia and 64 mm (2.10 mm) high.

The chemical process under study was direct reduction of lunar simulants, e.g., JSC-1 and MLS-1A, with carbon. In order to select a suitable container material in which to reduce the lunar simulants, five crucible material candidates, i.e., alumina, magnesia, yttria stabilized zirconia (YSZ),

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yttria, and tantalum, were tested. In all of the experiments, the oxide samples, i.e., JSC-1, MLS-1A, grunerite, and ilmenite, and their respective stoichiometric amounts of carbon were weighed and charged into their respective crucibles where they were well mixed. After mixing, the crucibles containing the reactants were placed on a graphite support tray in the HTF hot zone. The crucibles were separated from the graphite support tray with alumina wafers and shims to eliminate interaction of the crucibles with the graphite. After loading the crucibles, an argon purge was used at ambient to remove all of the air from the HTF chamber. The HTF power was turned on and the temperature was raised according to predetermined programs, e.g., a heating and cooling rate of 2.5 C/min. An argon flow rate of 100 to 142 scc/min was maintained throughout each experiment from the start of heating to cooling to room temperature. The reduction reactions were allowed to proceed at a maximum reaction temperature of 1660 ± 5 C (3020 ± 9 F) for periods as long as 23 hr. The experimental data included furnace temperature, product gas flow rate, and carbon monoxide QMS peak height. In addition, the power output variables and controller thermocouple readings were recorded. After the completion of each experiment, the crucibles containing the reduction products were visually examined, weighed and photographed immediately. Elemental identifications of the solid products were carried out using a computer aided scanning electron microscope (SEM) with energy dispersive x-rays (EDX) in which SEM photomicrographs and EDX spectra were regularly taken for the various solid phases. Phase identifications were performed using x-ray powder diffraction techniques in which diffraction patterns for various phases were obtained.

Two lunar simulants provided by NASA, i.e., JSC-1 and MLS-1A, ilmenite, and synthetic grunerite were reduced with carbon, i.e., graphite, in five candidate crucible materials, i.e., alumina, magnesia, yttria, YSZ (8 wt%) and tantalum carbide in various combinations. From the results of these Carbothermal Reduction Experiments, it was concluded the reduction of the lunar simulants with carbon was achieved as evidenced by the formation of carbon monoxide (QMS analysis), and iron silicide (SEM/EDX and X-ray analyses), thereby confirming the earlier Aerojet research. Reduction of the ferrous/ferric oxides started at ca. 900 C, as evidenced by the formation of carbon monoxide, the evolution of which steadily increased to a first peak value at 1100 to 1200 C. Carbon monoxide evolution started to increase again at ca. 1475 C. This increase may have resulted from the formation of a liquid phase which enhances the carbon diffusion process which, in turn, permits the reduction of the last of the iron oxides to occur, or the reduction of silica, or both. The reduction reaction rate increases steady to a second peak value at ca. 1600 C due to the reduction of silica, followed by formation of liquid metallic ferrosilicon. The reduction of silica is aided by the presence of liquid iron which forms a solution with silicon, thereby lowering its thermodynamic activity which, in turn, facilitates silica reduction. The results of the experiments indicate the temperature required for the complete reduction of lunar simulants mechanically mixed with carbon at a reasonable rate is ca. 1650 C with a total reaction time in excess of 20 hr.

The results of the experiments showed that, except for yttria and YSZ, all of the crucible materials were unstable with respect to the environment under which the reduction reactions were conducted. The YSZ crucibles survived under proper heating/cooling conditions, e.g., 2.5 C/min. However, these crucibles reacted significantly with the components of the reduction reactions. The YSZ crucible walls formed a thick diffusion layer with the metallic ferrosilicon.

The 100% dense yttria crucibles survived the reduction environments under proper heating/cooling conditions, e.g., 1.5 C/min. In sharp contrast with the YSZ crucible results, interface studies of the reduction reaction products with the yttria crucible walls showed that a thin interfacial product layer formed at the relatively higher reaction temperatures for relatively longer period of time, indicating the kinetics of such interfacial reactions are relatively slow. These results, in turn, strongly indicated that 100% dense yttria provides a suitable container for the Carbothermal Reduction of lunar simulants and regolith.