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A Large-scale Laboratory Demonstrator for the Production of Oxygen from Lunar Regolith: Challenges and Opportunities

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Abstract

Reduction of lunar minerals by hydrogen gas generates water. This principle has been the most long-standing idea to extract oxygen from the materials of the lunar surface. The ALCHEMIST-ED technology demonstrator serves to explore this process in a laboratory environment on a semi-industrial scale. We introduce the lunar geological baselines, give an overview of the system and discuss the challenges associated with the process. The learning opportunities and technological baselines arising from the planned studies will enable us to assess and advance the technology against competing approaches.

1. Introduction

The production of oxygen and metals from the lunar regolith has been the focus of significant and growing research interest over the last 20 years. In-situ pro-duction of water and oxygen is seen as being the first critical step towards sustainable space travel, reducing the reliance on materials from Earth. Here, we focus on hydrogen reduction of lunar minerals to extract oxygen and produce metals and will update on ESRIC's current work and research objectives. Reduction of the lunar regolith using hydrogen is one of several technologies proposed to extract oxygen bound in the minerals that make up the lunar surface – in this case mainly from ilmenite, $(Fe,Mg)TiO_3$ [e.g, 1,2]. While it is not new, it has fallen out of favour in comparison with technologies that potentially produce higher oxygen yields, such as molten salt or molten regolith electrolysis. Despite its low yields with raw regolith (typically about 1 % w/w O_2), it continues to be of interest due to its relative simplicity and the potential to enhance yield via regolith beneficiation. ALCHEMIST-ED is a large-scale demonstrator of a hydrogen reduction system designed and build by Space Applications Services and Metso:Outotec [3] under contract with ESA and currently being commissioned at ESRIC. The TRL-4 system accepts up to 1.4 kg solid mass charge and utilizes an externally heated fluidized bed reactor coupled to gas recycling. Following initial commissioning tests, the ALCHEMIST-ED will be used to enhance our understanding of this system in order to improve extraction efficiency and to optimize the end-to-end process.

1.1 Lunar geological baselines

Ilmenite is an oxide mineral with the ideal composition $FeTiO_3$ that occurs in various lunar mare basalt types, with the highest concentrations in high-Ti basalts. It forms as interstitial grains or as exsolution lamellae within pyroxene or olivine (Ca-Mg-Fe silicates). Lunar ilmenite differs from terrestrial ilmenite in its lack of ferric iron (Fe^{3+}) through solid solution with a hematite (Fe_2O_3) component and its enrichment in Mg through usually large proportions of the geikielite ($MgTiO_3$) solid-solution endmember. The abundance of lunar ilmenite varies from region to region, but it is estimated to be around 10-15 wt% of the lunar regolith in high-Ti mare areas. Such areas would be the prime target for economic utilization of ilmenite to produce oxygen via the reduction by hydrogen and subsequent water splitting.
Lunar ilmenite is a potential resource for in situ oxygen production, as it can be reduced by hydrogen or methane to yield metallic iron and titanium oxides, along with water or carbon monoxide and hydrogen, respectively. The reduction by hydrogen proceeds through the following simplified reaction:

$$FeTiO_3 + H_2 \rightleftharpoons Fe + TiO_2 + H_2O$$

The methane-based, carbothermal process can be simplified as the following reaction:

$$FeTiO_3 + CH_4 \rightleftharpoons Fe + TiO_2 + CO + 2H_2$$

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The carbothermal pathway is problematic under lunar conditions because carbon is a geochemically highly limited resource on the lunar surface and needs to be imported at high cost per mass. Also, hydrogen is a limited resource on the lunar surface and needs to be imported from Earth unless lunar polar ice deposits can be utilized. However, due to the smaller atomic mass of hydrogen, the cost for importing a stoichiometrically equivalent amount of hydrogen is considerably lower.

1.2 The ALCHEMIST-ED demonstrator

The ALCHEMIST-ED demonstrator (Fig. 1, 2) is a fluidized bed reactor with an internal volume of approx. 7.5 L and a nominal operating pressure up to 3.5 bar(g). The process gas is cycled by two parallel membrane pumps through the reactor, a cyclone dust separator, a dust filter, a condenser and finally through a preheater/heat exchanger back into the fluidized bed reactor. The preheater serves to transfer heat from the effluent H_2O-bearing gas to the recycled H_2O-poor gas coming from the condenser. The condenser currently operates down to temperatures of -10°C with a water-ethylene glycol coolant. ALCHEMIST-ED is extensively equipped with pressure, flow and temperatures sensors as well as an online gas analyser, all of which are providing data to the central PLC for continuous data logging during operation.

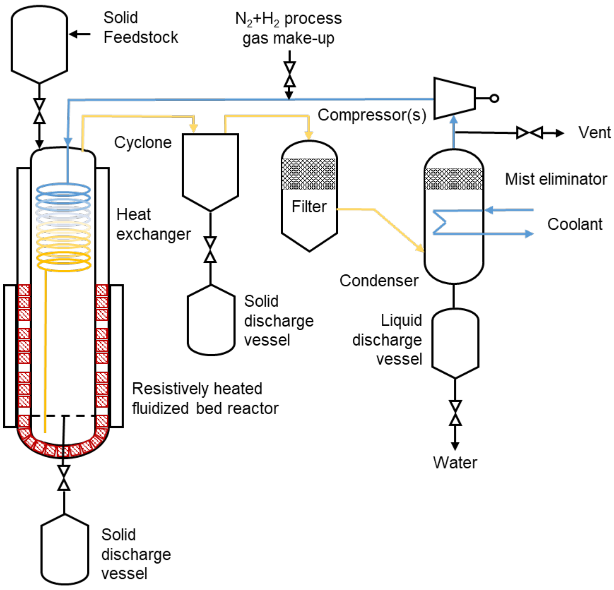

Figure 1: Schematic system diagram of the ALCHEMIST-ED demonstrator.

Figure 2: Physical characteristics of the ALCHEMIST-ED demonstrator.

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2. Technological Challenges

The near-term technological challenges are to assess and adapt the technology tested in ALCHEMIST-ED for lunar operation and flight-readiness. ALCHEMIST-ED serves to provide technological baselines to assess reduction by hydrogen against competing technologies, such as molten-salt or molten-rock electrolysis and provide planning for further development of the technology itself through innovative opportunities. We have identified several key research questions that need to be addressed to generate the required fundamental, technological, and economical understanding for further development. The order presented here does neither imply any particular ranking in relevance or prioritization nor does it insist on completeness.

2.1 Thermodynamics of the reduction

Reaction (1) constitutes a forward and reverse reaction. The rate of the forward reaction progress ξ is limited by the reverse reaction with water in the gas. Because under equilibrium conditions both rate are equal, the system has to be driven away from equilibrium by removing water from the cycled gas stream. The reaction rate $d\xi/dt$ in such a case is primarily controlled by the difference between the actual and the equilibrium water partial pressure in the gas (linear kinetics):

$$\frac{d\xi}{dt} \propto \left(p_{H2O}^{eq} - p_{H2O}\right)$$

The influence of water separation on the chemical potential of the gas driving the forward reduction needs to be understood, i.e., high H_2O/H_2 ratios (high fO_2) drive the reaction away from reduction and, hence, water needs to be removed as efficiently as needed through the condenser system and/or adsorption.

2.2 Reaction pathways and kinetics

Depending on the thermodynamics, temperature, and microstructures of the solid products, the reduction rates may vary over orders of magnitude. Proper reaction progress models (i.e., core-shell geometries, anomalous diffusion, rate models) and size dependencies need to be established. The evolution of solid reaction products on the ilmenite feedstock is expected to be very complex, involving a heterogeneous phase assemblage (metallic Fe, Mg titanate residuals, non-stoichiometric Ti oxides; [4]) and multiple mobile species diffusing through the reaction products (e.g., Fe^{2+}, O^{2-}, e^-, H species, vacancies). If the reactions are controlled by diffusive transport, then $d\xi/dt$ is a function of time (parabolic kinetics):

$$\frac{d\xi}{dt} \propto \frac{1}{\sqrt{t}}$$

While bulk diffusion through product phases is slow, anomalous diffusion along grain boundaries of the heterogeneous products and through voids in the product layer can be orders of magnitude faster. Hence, the textural evolution of grain sizes and porosity is and important controlling factor of reaction kinetics and eventually the power required to drive the process.

2.3 Energy and power demands

The reduction process needs to be optimized with respect to duration and extent of the reduction reaction. Here, the understanding of reaction kinetics and the trade-off between increasing thermodynamic driving forces and investing in water separation efficiency are crucial. It is expected that the initial reaction progress is characterized by linear kinetics that then switches to parabolic kinetics once the bottleneck of diffusion-limited transport is reached (Fig. 3).
The objective of process optimization is to maintain fast linear reaction kinetics to the largest reaction extent and longest time possible. This requires understanding the physicochemical controls on the textural evolution of the reaction products and the available means to manipulate it for a maximum of fast, anomalous diffusive transport. Feedstock particle size, particle shape, surface abrasion and reaction temperature are expected key factors controlling the course of the reaction. While higher temperatures would increase rates, this factor has the largest impact on the power demands of the system and, hence, needs specific attention.
Once parabolic kinetics have set in, the reaction rates $d\xi/dt$ will drop considerably and reaction to completeness ($\xi = 1$) will take much longer than in the linear case. As this in inevitable, a criterion to stop the reaction before completion

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needs to be derived as a prolonged reaction, and therefore energy input for comparably low reaction progress, would become economically prohibitive.

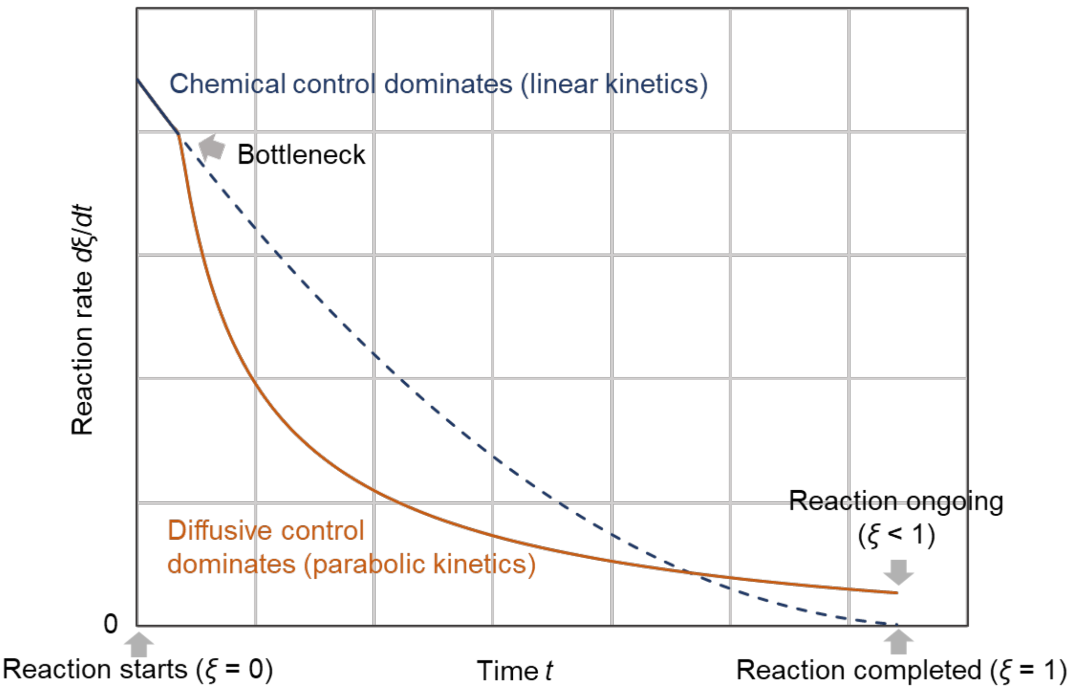

Figure 3: Schematic reaction rate-time dependency for the reaction of a spherical particle (although linear kinetics are not time dependent, the spherical geometry imposed a decrease in reaction rates over time).

2.4 Beneficiation of the feedstock

Because ilmenite is the main mineral that can be reduced by hydrogen, its low modal (vol%) abundance and therefore oxygen yield with respect to raw regolith requires enrichment of ilmenite through size- and material-selective separation. The physical beneficiation of the feedstock therefore is crucial, and solid understanding of the separation of the target mineral ilmenite under challenging conditions needs to be obtained. Industrial beneficiation in terrestrial mineral processing commonly relies on copious amounts of water, which is not going to be available in a lunar setting. Alternative methods of mineral separation, for example driven by differential electrostatic and magnetic responses and contrasting densities, need to be understood and developed. Although this is not in the functional scope of the ALCHEMIST-ED demonstrator, efficient beneficiation technologies are inherently linked to the process optimization as the heating of non-reactive components of the regolith feedstock would easily become prohibitive in terms of power demand.

2.5 Fidelity of regolith simulants

Technological knowledge at low TRL requires extrapolation and de-risking to transfer it to the lunar environment. Regolith simulants may contain undesired hydrous minerals that release water during the high-temperature processing and bias the understanding of water production through reduction [3]. Ferric minerals absent on the Moon but present in terrestrial simulants, e.g., magnetite (Fe_3O_4) or hematite (Fe_2O_3) exsolution in ilmenite, are of additional concern, because their reduction to may lead to overestimation of reaction rates and yields. In particular, hematite is a common mineral that formed from the high-temperature $FeTiO_3$-Fe_2O_3 solid solution during cooling. Its exsolution lamellae may be present a nanometer scales and may not be readily detected by standard microscopic techniques. The presence of such ferric iron impurities is expected to alter the reaction kinetics by having an influence on the thermodynamical driving force (Fe_2O_3 is easier to reduce than $FeTiO_3$) and the textural evolution of the product layer. Moreover, despite the fact that hematite is just a weakly magnetic (canted antiferromagnet) material, the lamellar intergrowth of hematite and ilmenite at nanoscales leads to exotic magnetic properties [5] that may impact the transfer of magnetic beneficiation from terrestrial simulants to actual lunar ilmenite, where such exsolution features are absent due to the highly reduced petrology of the lunar source rocks.

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2.6 Continuous operation

At present, ALCHEMIST-ED is operated in batch mode with feedstock masses up to 1.4 kg. Initial provisions have been made to be able to move to semi-continuous operation through using lock hoppers and valves gating the reactor from the feeding and discharge vessels. Challenges in this respect are the flowability of material through the feeding system, in particular considering the lower gravity on the lunar surface, the wear of valves in the presence of mineral dust and the retention of hydrogen in the system. As hydrogen is a highly limited lunar resources, technological development effort is needed to ascertain a minimum of loss during it cyclic utilization in the process. This not only applies to the reduction process studies in ALCHEMIST-ED but also to the subsequent water splitting through electrolysis or by other means.

2.7 Qualitative and quantitative impurity assessment

The utilization of water/oxygen extracted during the reduction process imposes purity constraints on the final product, e.g., for electrolysis. Due to the heterogeneous nature of the feedstock, contaminants are generated in the process, e.g., H_2S from troilite, FeS, or HCl from chlorapatite, $Ca_5(PO_4)_3Cl$. Understanding of the reactions and the contaminant levels in the product are required to optimize the process, e.g., through pre-reduction beneficiation and post-reduction purification. Beneficiation would selectively remove the sources of unwanted reaction products to minimize the contaminant load in the output stream and to reduce corrosion of the system itself. Purification of the effluent gas stream aims at removing unwanted reaction product or selectively recovering valuable by-products for downstream use in other ISRU processes.

2.8 Separation and utilization of by-products

Metallic iron and other solid by-products (e.g., Ti oxides) are of interest for downstream utilization. They need to be separated and refined to interface with the relevant technologies, e.g., metal 3D printing. Currently, this is studied at ESRIC also for metal separates obtained from FFC molten salt regolith reduction, which will allow a better understanding of the potential trade-offs between electrochemical reduction and the reduction by hydrogen.

3. Opportunities

The research utilization of ALCHEMIST-ED addresses, directly or indirectly, all of the challenges outlined in the previous sections. It provides the opportunity to generate well-constrained technology baselines for further development and assessment of the technology for lunar operation. Reduction by hydrogen has been the longest standing concept of oxygen production on the Moon and potential improvements, such as using neutral atomic hydrogen for the reduction instead of molecular di-hydrogen, could drastically enhance efficiencies and yields. The lessons learned from the utilization of ALCHEMIST-ED will be essential to derive concepts and designs for flight hardware. Ongoing multicriteria trade-off assessment will eventually identify the technology most suitable for sustained and sustainable lunar operation.

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