Design and Test of a Concentrated Solar Powered Fluidized Bed Reactor for Ilmenite Reduction

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Abstract. This paper describes a solar powered fluidized bed reactor for the reduction of ilmenite ($FeTiO_3$) with hydrogen (H_2) at up to 950°C. It is intended to be a terrestrial demonstrator of this technology for the use on the Moon for extraction of oxygen from lunar soil (regolith). The goal of the reactor development was to build a full scale reactor including all necessary peripheral components to test the reaction on Earth, and to demonstrate solutions for many of the challenges it will be facing on the Moon.

The center-piece of the reactor is a fluidized bed with a capacity of 25kg of ilmenite. The concentrated solar power enters the reactor vertically from the top through a quartz window, allowing direct heating of the particles without the need of a heat exchanger wall. Feeding and removal of the solids is done in continuous mode by auxiliary fluidized pipes. The gas supply includes a recirculation pump, flow controllers, and an electrolyzer. The off-gas treatment includes cooling, hot gas cleaning from remaining fines, and active separation of the desired product water from the gas stream. The reactor was extensively tested in the first half of 2017 in the 60kW Solar Furnace at the Plataforma Solar de Almería (PSA) within the first solar test campaign of the Oresol-project.

INTRODUCTION

One of the major challenges of future spaceflight beyond low Earth orbit is the supply of spacecraft and crews with vital resources, like water, oxygen, and rocket fuel. The in-situ production of some resources on the Moon, Mars, or asteroids can significantly reduce the amount of mass needed to be launched from Earth.

The most needed and at the same time most abundant resource on the Moon is oxygen. The main problem is that oxygen release from the regolith requires high temperatures due to the strong chemical bonds in the minerals. The process with the most benign operation conditions is hydrogen reduction of the titanium-iron oxide mineral ilmenite (eq. (1)) at 800 - 1000°C, and subsequent water electrolysis (eq. (2)) [1]:

$$FeTiO_3 + H_2 + \textit{heat} \rightarrow Fe + TiO_2 + H_2O \quad (1)$$
$$H_2O + \textit{el.} \rightarrow H_2 + \tfrac{1}{2}O_2 \quad (2)$$

While the hydrogen from reaction (2) is returned to reaction (1), the oxygen is the desired product.

Nowadays, photovoltaics is by far the most used power source in spaceflight in the inner solar system (besides chemical propulsion of course). Reasons are that the main power demand is for electricity, the very predictable availability of solar radiation, and its simplicity (no moving parts and no need to reject surplus heat).

On Earth, concentrated solar power is a well-established technology in the field of electricity production. In other energy consuming sectors, like e.g. the high temperature process technology, there is still a lot more need of technological development, especially if the receiver and the reactor are combined into one unit. Key to success is the adaption of the reactor to the special requirements of concentrated solar power. As a reactor for the reaction (1) needs mainly thermal power, and solar radiation is abundantly available on the surface of the Moon, it makes sense to develop a solar thermal reactor for this application.

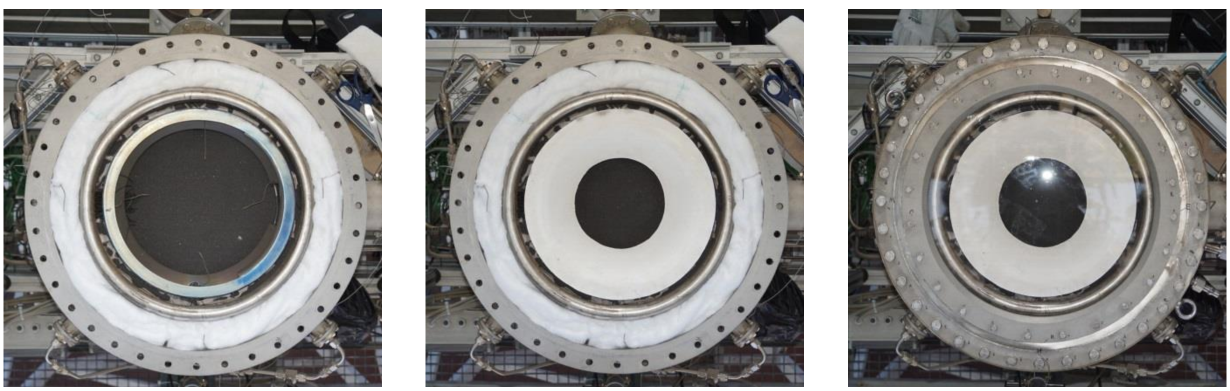
SYSTEM DESIGN

A solar powered chemical reactor for the lunar ilmenite-hydrogen reaction (and generally for whatever slow-kinetics reaction of particulate solids with a gas) must meet several requirements. They are continuous operation due to the nature of solar radiation, the ability to heat and process large amounts of solids in particle form, a solids residence time of several (tens of) minutes, and a good mixture of the solids with the gas reagent. All these conditions can be satisfied with a low expansion fluidized bed reactor.

Fluidized beds have some very remarkable properties [2]. One is their liquid like behaviour. They form a horizontal surface, can flow like a liquid between two vessels, and equalize their levels. Another one is their very uniform temperature distribution. A disturbance is generally equalized within few minutes. Efficient heat transport and heat transfer avoids problems with local hot spots. These properties allow continuous, automatically controlled operations with easy handling. An important disadvantage is the wide residence time distribution of the solids that limits their chemical conversion to somewhat below 100%.

A low expansion fluidized bed reactor is basically a well-insulated, cylindrical vessel, filled with the solid particles. It has a specially designed floor, the gas distributor, which allows the gas to enter from the bottom in a homogeneous way, but avoids the particles to leak into the distributor area.

The concentrated solar radiation enters the reactor (Fig. 1) vertically from the top through a flat quartz window. The pathway for the light has a conical shape with the narrowest point, the aperture (ø150mm), about 50mm above the surface of the fluidized bed. An additional gas stream injected radially between the window and the aperture avoids the deposition of fines from the bed on the inside of the window.



(a) (b) (c)

FIGURE 1. Solar fluidized bed reactor view from top. (a) Bed surface. (b) Aperture. (c) Window.

The continuous supply of the particles (Fig. 2a) starts in a funnel located behind and above the reactor. From there, the particles flow down the so-called standpipe. Their mass flow is measured by a balance below the funnel and controlled by an auxiliary small gas stream that partially fluidizes the standpipe, a configuration generally known as “L-Valve”. After the L-Valve, the particles enter the riser, a well fluidized vertical pipe, where they move up again and then simply spill into the reactor.

The removal of the particles (Fig. 2b) is done by a simple overflow that at the same time defines the bed level in the reactor. From there, a pipe goes out with the shape of a fluidized syphon. This allows to compensate for the slight over-pressure in the reactor, excludes that atmospheric air accidentally enters from outside into the reactor, and especially avoids that the fluidization and product gases escape through the particles out-pipe.

The most complex part of the peripheral components is the so-called “downstream section”, where the treatment of the gas from the reactor is done (Fig. 3). In the freeboard of the reactor, a specially designed gas outlet ring (see Fig. 1a) collects the gas but allows the entrained particles to precipitate back into the bed before leaving. Once out of the reactor, the gas needs a multiple treatment. Besides being very hot, it contains excess fluidization gas, the product water, some particles from the bed that escaped despite of the outlet ring, and some possible substances from chemical side reactions. First step is a moderate cooling to a temperature well above 100°C to avoid a premature condensation of the product water. The second step is the cleaning of the gas from remaining particles by the particles separator. This device takes advantage of the inertia of the particles in the gas stream suddenly changing direction by 180°, followed by a filter for the remaining fines. In the subsequent second cooler, the now clean but still hot gas is cooled down to near ambient temperature. In this step, a major part of the product water is condensed. To extract as much water as possible, the gas stream is cooled several degrees below ambient in the following water separator with the help of a Peltier-element. This decreases even further the ability of the gas to carry water, hence releasing the maximum possible amount of water from the system. The water accumulates on the bottom of the water separator and is extracted from there by a small peristaltic pump. The reasons are the same as for the syphon of the particles outlet, to avoid escape of the gas and erroneous entry of atmospheric air. Furthermore, this device allows the measurement of the produced water volume. The sub-cooled gas, now with a relative humidity of 100%, is guided back again into the cooler, now working as a "warmer", increasing slightly again the temperature of the gas back to about ambient. This measure is important because it reduces notably the relative humidity of the gas, hence reducing subsequent condensation in the piping. Finally, the now "cool, clean, and dry" gas partially (depending on the total gas flow) leaves the system through a check-valve with the task to control the pressure, and partially (at nominal operation conditions by far the major part) is returned into the upstream (flow-control) section by the recirculation pump.

FIGURE 2. Continuous particles supply (a) and removal (b).

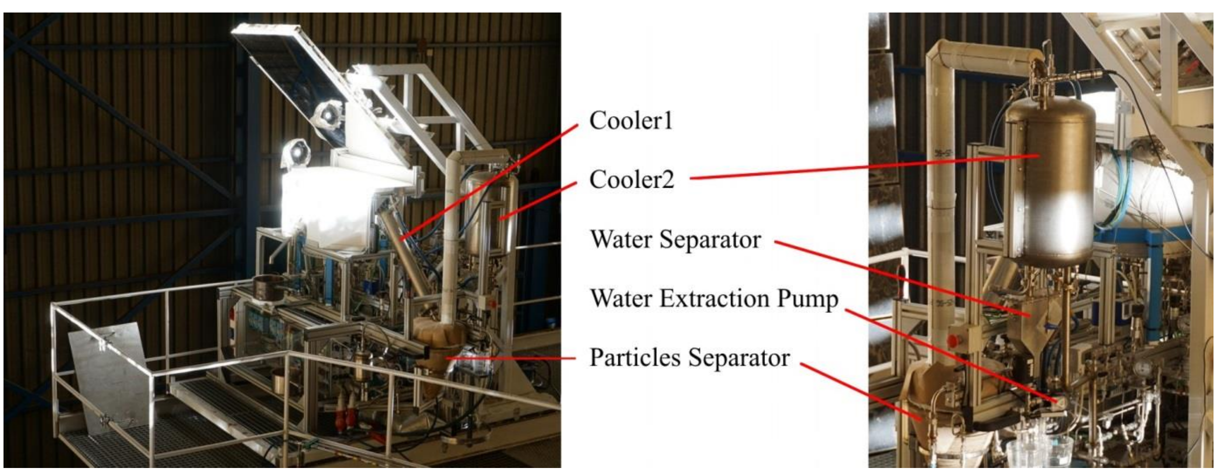


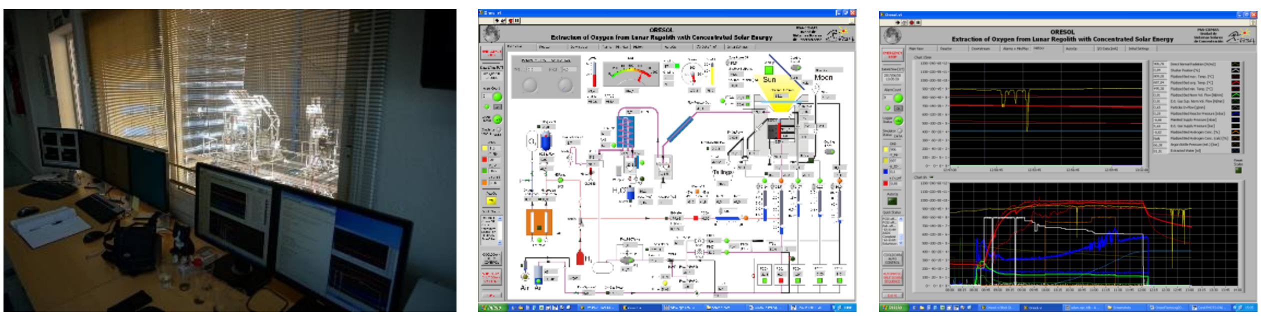
FIGURE 3. Downstream section (off-gas treatment).

TEST OPERATION

Data Acquisition and Control

The Oresol system counts with a total of 80 thermocouples, 6 flow controllers, 2 additional flow transmitter, 2 weight sensors (strain gauges), 14 pressure transmitter, 1 hydrogen sensor, 3 (water) level indicators, and 7 digital outputs to control (besides the 6 flow controllers) 3 pumps (main gas, hydrogen, and water extraction), 2 fans, the electrolyzer, and the Peltier cooler. Furthermore, some data from the PSA Solar Furnace environment is fed into the system, like solar radiation (DNI), weather data (temperature, wind), and the position of the shutter. To get a good resolution of the temperature distribution within the fluidized bed, there are 15 thermocouples submerged in the bed, arranged in three levels in horizontal crosses of five.

The SCADA (Fig. 4) is programmed in LabVIEW. Besides the typical features like visualization and logging of the data, it allows for a large amount of automated operation. This includes the automatic setting of four gas flows (main bed gas, window gas, particles in- and outflow pipe fluidization) in dependence of the corresponding temperatures, the automatic (and somewhat problematic) control of the particles inflow through the L-Valve, the hydrogen feeding, and the Peltier cooler. If nothing goes off-nominal, only the position of the shutter has to be adjusted manually by the operator. Further functions are alarms for high (or low) temperatures or pressures, low gas or particles supply, automatic cool down and shut down procedures, and many more. A graphical representation of the most important data of the last 15 minutes and the last 6 hours allows immediate short term and long term assessment. A simulation mode for testing of new features and training of the operator completes the program.



(a) (b) (c)

FIGURE 4. Data acquisition and control.

Operation Strategies

Regarding the gas supply of the fluidized bed reactor, two tasks of the gas must be clearly distinguished. At one hand side, it has to perform the chemical reaction, on the other hand side, it is needed for fluidization. The latter one determines the total flow rate. To match both tasks, the reactor geometry must be chosen adequately.

The ultimate goal of testing is operation with a supply of 100% hydrogen in the system. However, this option was postponed already in an early stage of the project due to safety concerns. In the meantime, the operation was done with argon for fluidization and only a small amount of hydrogen (<10%) for the chemical reaction. Still another step before, the system was operated with atmospheric air, without chemical reaction, and the temperature in the bed was limited to 400°C to avoid damage on graphite parts.

The goals of the five tests of this “Phase 1” were to learn to know in depth the behavior of the fluidized bed with the particles, to check the proper functioning of most of the peripheral components, to hone the data acquisition and control program, to confirm the initial predictions of the gas demand of the bed in function of the temperature, and finally to establish efficient procedures to pass as fast as possible through the low temperature part of the heating process. The latter one is of special practical importance because this is where the gas consumption is highest, and hence determining the cost of the tests when argon is used.

The six tests of “Phase 2”, with pure argon, were to determine the operation parameters (temperatures, gas flows and pressures, solar power), and to demonstrate the ability of the system to reach the minimum nominal operation temperature of 800°C. Additional tests helped to understand the behavior of the system with continuous particles in- and outflow, with a special interest on the variation of the solar power demand.

Finally, in "Phase 3", hydrogen was added to the system and the temperature further increased. Fifteen tests were performed, eleven of them with hydrogen, two for particle flow / solar power tests, and two for TV interviews. The basic goal here was to demonstrate that the reaction really occurs. Further objectives were to gain initial information about the water quality, and, as a bonus, to calculate the hydrogen conversion rate. It must be emphasized that in this test campaign, the operation was just opposite to how it will be in the final application. While on the Moon the operation would be done with a considerable excess of hydrogen (factor 4 or so) and a rather low concentration of ilmenite in the particles (5-25%), the testing in this campaign was with a huge excess of ilmenite and low hydrogen. Hence any quantitative analysis of the chemistry has yet only limited meaning.

Particles

Obviously it is not possible to perform the tests with real material from the Moon. To overcome this problem, several manufacturers provide so-called lunar soil simulants; [3] gives an overview. One of the best known simulants is JSC-1A produced for NASA, and also purchased for the Oresol project. However, as this simulant represents Ti-poor lunar highland soil, it can only be used for fluidization studies, but it's not suitable for chemical tests of ilmenite reduction. Therefore the particles used in this testing campaign were pure ilmenite, with a total of one metric ton obtained from Tioxide (Spain), originating from Mozambique.

They have a rather narrow grain size distribution averaging around 150µm, putting them into the Group B of the Geldart classification [4]. For future, more realistic and optimized testing, another 200kg were purchased, with a medium grain size of 48µm, putting them into the more preferable Geldart Group A. The particles of this group tend to fluidize rather smoothly with only small gas bubbles, while the bubbles in a bed made of group B particles tend to grow to big sizes, making the surface of the bed rather agitated, and worsening the contact between the gas and the solids. Mixing the smaller ilmenite with a properly screened fraction of JSC-1A should eventually give a simulant with sufficiently adequate properties for testing.

RESULTS

The Oresol-Spring-2017 test-campaign started in December 2016 and lasted (with interruptions) until the end of June 2017. All three primary goals of the campaign were successfully achieved:

1. Identify the gas flow demand of the main fluidized bed in the reactor as a function of the temperature.
2. Operate the reactor at >800°C solely heated with concentrated solar power.
3. Demonstrate water production from the reaction of the ilmenite with hydrogen.

Further (secondary) goals like the demonstration and control of the continuous particle feed / removal from the reactor, or the off-gas treatment were also accomplished.

Fluidization Gas Demand

The gas demand at minimum fluidization can be approximated for the Oresol system by a $\dot{V}_N \sim 1/T^{1.7}$ curve ($\dot{V}_N$… gas volume flow at norm conditions). Several measurements were done to verify the prediction (Fig. 5). To get a meaningful turbulence and mixture in the bed, additional gas must be supplied (factor u/u_{mf}). The assessment of the fluidization quality was done during the solar tests through visual observation on a TV-monitor. During Phase 1 and especially the first two tests of Phase 2 reaching 700°C, it turned out that u/u_{mf} had to be increased continuously during operation with increasing bed temperature. Analysis of the manual adjustments (red in Fig. 5) during the test on Feb 1st led to a new correlation, with the flow following a $\dot{V}_N \sim 1/T$ curve (blue in Fig. 5). It was included into the control program under the label "empiric" and worked perfectly for the remainder of the tests.

Power Balance

To properly design the concentrator system for future application, it's essential to break down the different heat sinks of the reactor. Energy is needed for:

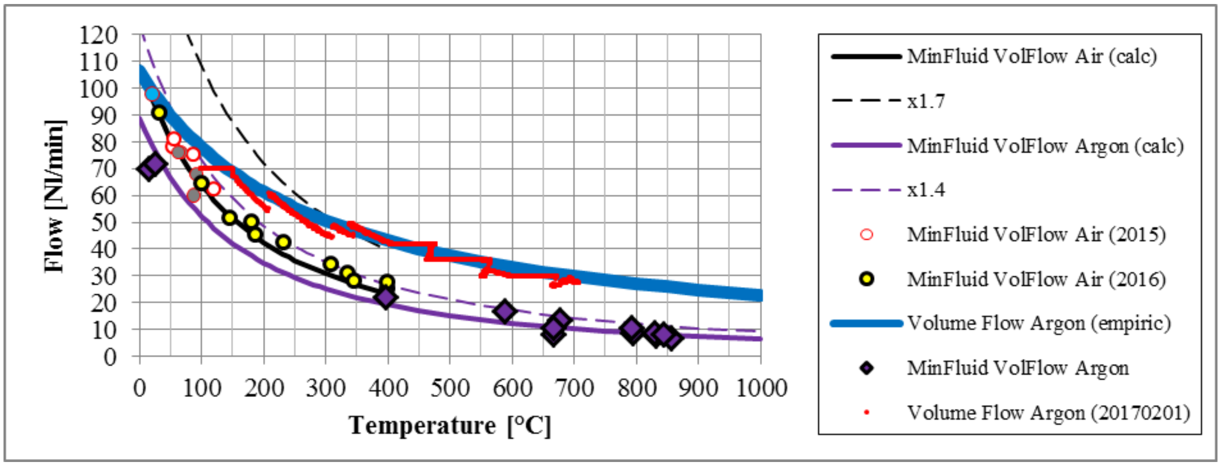


FIGURE 5. Norm volume flow vs. temperature. Calculated and measured data.

- Temperature change of the particles actually present in the bed. This happens during unsteady operation like heat up at the beginning of the test or cool down at the end.
- The heating of the particles entering the bed. This happens also during steady-state operation.
- The heating of all gas flows entering the reactor. They do not necessarily reach all the same temperature (e.g. window gas remains cooler than the main bed gas), but are reasonably averaged in the gas outlet.
- Infrared radiation losses. They are described by the Stefan-Boltzmann T^4-law. As no data for the real bed surface temperature is available, the upper layer average temperature was taken as reference. With the diameter of the hole in the alumina protection above the window taken for the emitting surface, and the emittance set equal one, the best match with the measured data was obtained (see below).
- Conduction through the reactor walls and the window. Neglected due to low contribution.
- Temperature change of the fluidized bed container walls and adjacent insulation. This heat sink was also neglected due to the extreme difficulty of proper modelling. It's notable during phases of rapid temperature change (see below).
- The chemical reaction. With activation energy of 93.4kJ/mol [5], its contribution is below 0.2kW in these tests and is neglected as well.

Unfortunately, there was no possibility for the measurement of the incoming (solar) power. Its calculation however seems straightforward, only the DNI, the shutter position, and the nominal power of the solar furnace (60kW at 1000W/m^2 and 100% shutter opening) have to be multiplied. The problem is that the latter one is only valid for an unobstructed, horizontal beam and a very clean heliostat with no cosine losses. In the case of Oresol, there are several additional reductions, namely the partial blocking of the incoming (horizontal) ray by the alumina protection of the reactor, the intercept of the diagonal mirror, its reflectance, and the intercept of the aperture. This reduction was determined by a power analysis. The values obtained varied from 0.65 to 0.43, depending on external conditions like hazy sky, the tracking precision, and/or the cleanliness of the concentrator and the heliostat.

The test from June 7th (Fig. 6) is particularly useful for a power balance, because it has unusually clean data for three different particle mass flows. Operation started around 08:30 (UTC) with the reactor still warm (240°C) from the operation the day before. Around 09:45, the nominal operation temperature was achieved and the incoming power reduced by somewhat closing the shutter. After some time for stabilization, particles inflow of 110g/min started at 10:30, lasting for one hour. Then, at 11:30, the inflow was doubled to 220g/min, until it was cut off at 12:00. The shutter was closed another half an hour later at 12:30, and the temperature started dropping. Finally, 15 minutes later, the gas flows were also turned off, finishing the test. Hydrogen was not used on this day. The following observations can be taken from Fig. 6:

- At the beginning, with the particles still cool, most power goes into heating of the bed (grey line).
- With increasing temperature (red), the IR-losses (orange) become substantial.
- Also there starts to be "missing" some power in the balance (blue line > 0). This can be well explained by heating of the reactor wall and the insulation not included in this balance.

- When reaching the operation temperature, most solar power is lost by IR. A small part is heating the gas (green), the balance (blue) is fulfilled again to a large extent.
- With the start of the particles inflow (black), also the solar power (yellow) is increased. The balance (blue) remains fulfilled.
- The same is valid when the particles inflow (black) is doubled. The increased fluctuations in the data are due to imperfect fluidization at some places caused by a non-uniform flow through the distributor.
- After turning off the particles flow (black) and adaption of the shutter position, most of the solar power is lost again by IR radiation (orange), and the balance (blue) continues to be fulfilled. The power balance is best for this test with a reduction factor of 0.49.
- When the shutter is closed, the bed cools rapidly due to IR. The power balance (blue) becomes slightly negative, because the reactor walls now try to keep the bed hot. This part of the data is of special importance to confirm the assumptions for the infrared losses, because it's free of the correction needed for the incoming solar radiation. Figure 7(a) shows the good fit between calculated (yellow) and measured (red) power during cooling between 850 and 700°C. Figure 7(b) shows the orange glow of the 900°C fluidized bed immediately after defocus. Despite of the fact that the particles never melted, the fluidization causes the beautiful impression of a liquid, strongly bubbling *solar powered lava lake*!
- When the gas flows are finally turned off, the cooling rate drops dramatically. This is because the heat transfer within the bed changes from convective to conductive. The most upper layer cools rapidly, leaving the interior of the bed well insulated at high temperature and making the used IR calculation invalid.

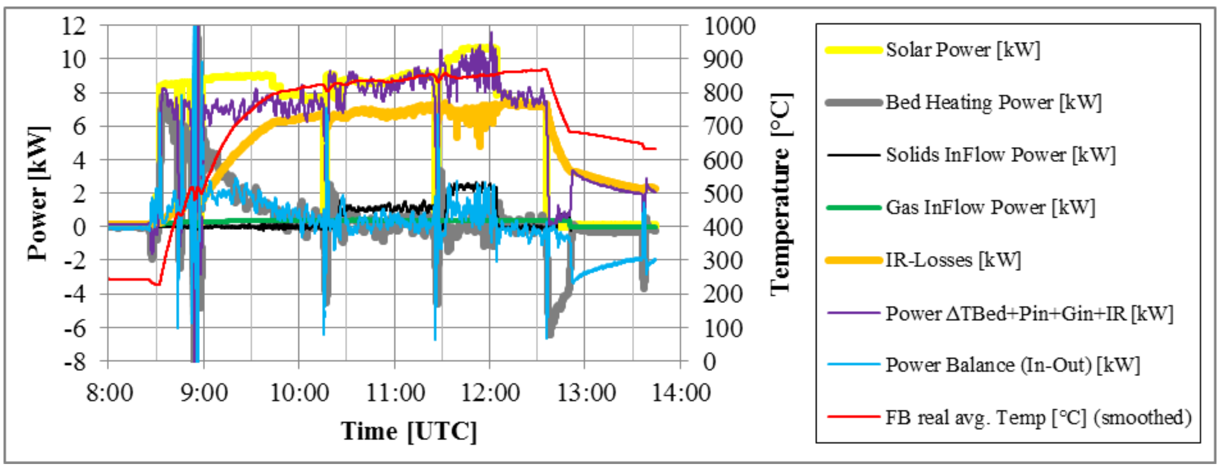
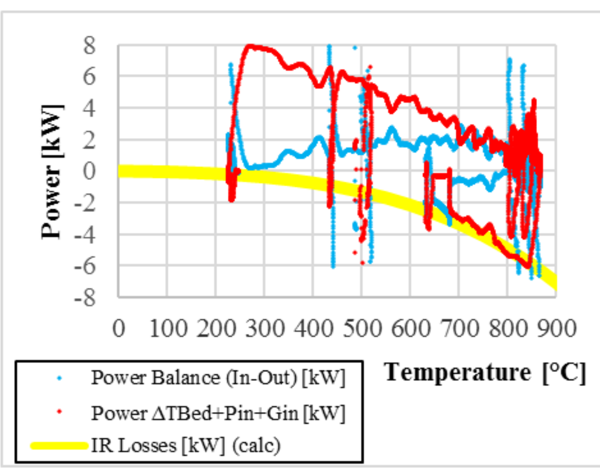


FIGURE 6. Power and temperature vs. time (test on June 7th).



(a)

(b)

FIGURE 7. (a) Power vs. avg. bed temperature (test on June 7th). (b) Glowing fluidized bed ("solar lava lake") at 900°C.

Several conclusions can be drawn from these observations. The infrared losses peaking 8kW are higher than expected. This can probably be improved by applying an insulation below the aperture cone, reducing this way the area of the hot surface. The power demand for the heating of the feed particles matches the calculations for pure ilmenite and is in the order of 12kW/(kg/min). The heating of the gas consumes less than 0.5kW and hence is only of minor importance. These data allow a rough first estimation for the concentrator size of a future plant on the Moon. With an (extraterrestrial) DNI of 1367W/m^2 and several further assumptions (reflectance, achievable ilmenite concentration, etc.), a concentrator diameter in the range of 4 to 5 meters seems reasonable.

Water Production

The final primary goal was the demonstration of the extraction of water from the ilmenite reduction. Tests with up to 10mol (224dm^3) corresponding to nearly two hours of hydrogen operation at 2Nl/min were done. An interesting observation was that there was a delay of several minutes between the hydrogen injection and the water release. The best conversion rate of 56% was achieved in the final test of the campaign. Next steps will include an increase of the range of the hydrogen flow controller and an improvement of the water separator unit.

SUMMARY AND OUTLOOK

A concentrated solar powered fluidized bed reactor for the reduction of ilmenite ($FeTiO_3$) with hydrogen (H_2) at up to 950°C was designed, built, and successfully tested. All primary testing objectives were accomplished. The basic operation parameters, especially the main fluidized bed gas flow, were determined. Continuous particle feed and removal was demonstrated. A maximum bed temperature of 977°C was achieved. The desired chemical reaction was carried out, converting up to 56% of the hydrogen into water with the oxygen being extracted from the ilmenite mineral. It arose during the tests that the system is also able to extract very efficiently the crystallization water in the minerals, thereby making it suitable for water extraction on Mars.

Nevertheless, many possible improvements were identified. For the short term, they include a higher hydrogen flow, a better water separation, a better gas distributor in the main fluidized bed, the installation of thermal insulation under the aperture cone, and the use of particles with a smaller grain size. Depending on the results, measures must be taken to improve the quality of the product water to make it suitable for the decomposition into hydrogen and oxygen in an electrolyzer. Another high priority task is the development of a lightweight foldable and/or inflatable concentrator system [6]. The achieved data suggest a diameter between 4 and 5 meter.

For the long term, the adaption to the lunar environment will be a major challenge. This includes vacuum of space, reduced lunar gravity, properties of the real lunar soil, and identification of ilmenite-rich spots on the Moon.

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