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Continued Development of an Automated Carbon Formation Reactor as a Life Support Technologies Solution for Long-Duration Manned Missions and In-Situ Resource Utilization

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Long-duration manned missions have a critical need for life support loop closure both for the journey, and for long-term survival in lunar and Martian surface colonies. The current technology is the Sabatier system, which has the potential for up to 50% oxygen recovery. This will not be sufficient to sustain manned flight beyond low Earth orbit. Therefore, this system must be greatly modified, or other life support technologies must be developed with a higher potential for oxygen recovery. These systems may take the form of a Series Bosch system that has the potential to achieve 100% oxygen recovery from metabolic carbon dioxide, or a carbothermal reduction process to extract oxygen from surface regolith. pH Matter's automated Carbon Formation Reactor (CFR) is a critical component of both systems, providing both solid carbon that can be used as a feedstock for carbothermal reduction as well as water, which can be electrolyzed to release oxygen. pH Matter's automated CFR has been demonstrated at the four-crew member scale at both ambient pressure and at an elevated pressure of 1379 kPa (200psia). The pressurized system was developed for use on the lunar or Martian surface for human-occupied habitats. The team will demonstrate the highly automated CFR reactor system at the forty-crew member ISRU-scale in mid-2024.

Nomenclature

CFR = Carbon Formation Reactor
RWGS = Reverse Water Gas Shift
CO = Carbon Monoxide
CO_2 = Carbon Dioxide
ISRU = In-Situ Resource Utilization
SLPM = Standard Liters per Minute
TRL = Technical Readiness Level
4-CM = Four Crew Member
MSFC = Marshall Space Flight Center
GRC = Glenn Research Center
SEM = Scanning Electron Microscope
PFD = Process Flow Diagram

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I. Introduction and Background of Technology

To facilitate extended-duration manned space travel and extra-Earth habitation, substantial improvements need to be made to life support technology to provide adequate oxygen for crew members. The current system that has been used on the International Space Station is the Sabatier system. This system creates methane as a byproduct, which is vented overboard. Due to the loss of carbon and hydrogen atoms, this system requires constant resupply of hydrogen and only has a 50% theoretical oxygen recovery potential[1]. This is not sustainable for missions far beyond low Earth orbit. One viable alternative is the Series Bosch system, which has the potential for 100% oxygen recovery. The byproducts from this system are water, which can be electrolyzed into hydrogen and water, and solid carbon, which can be used for a variety of in-situ purposes. pH Matter's Carbon Formation Reactor (CFR) is a critical component of this system. This system can be found below in Figure 1.

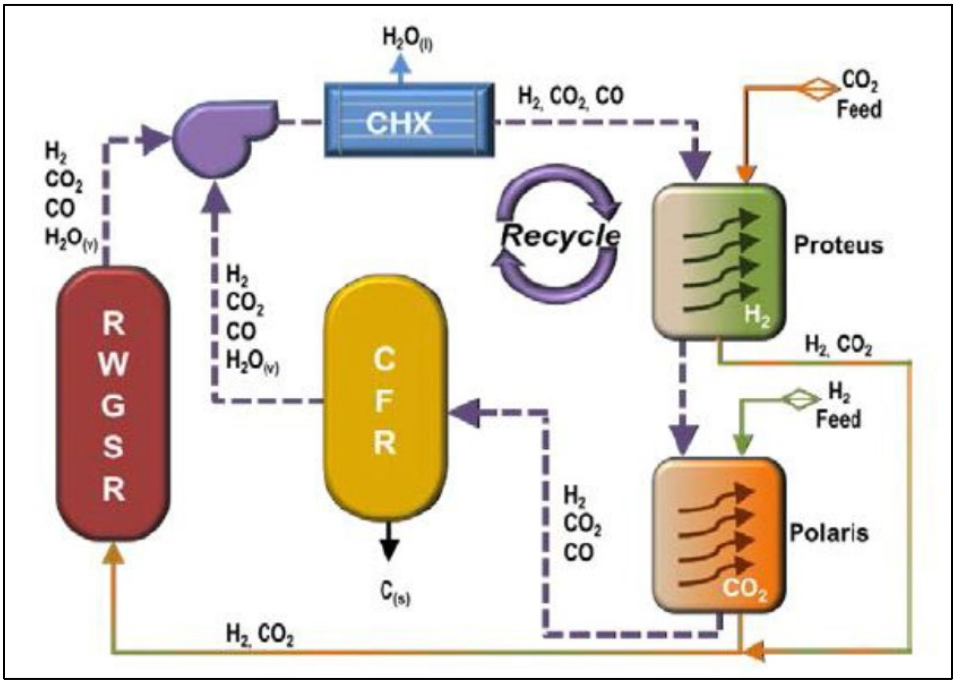

Figure 1. PFD of Integrated Series Bosch System in which the pH Matter CFR is Integral[2]

pH Matter's CFR converts carbon monoxide and hydrogen produced in the upstream reverse water gas shift (RWGS) reactor into solid carbon, water, and carbon dioxide using the Boudouard Reaction. The water produced is electrolyzed into hydrogen and oxygen, and the carbon can be used for a variety of in-situ purposes. Both the carbon dioxide and hydrogen return to the RWGS reactor with the incoming metabolic carbon dioxide from crew members, closing the gas loop. The potential for complete oxygen recovery from the system shows a clear advantage of the Series Bosch system over the Sabatier system due to the substantially lower resupply mass required.

The advantage of pH Matter's CFR over previous CFR iterations is the degree of automation and subsequent reduction in crew member interaction[5]. Astronauts onboard have many daily tasks to complete, so it is important to minimize the degree of crew member intervention in as many system processes as possible. Previous CFR prototypes have required extensive attendance to operating, in the form of manually delivering the catalyst, and the required adding and removing of coated substrates[2]. pH Matter's four crew member (4-CM) CFR reactor systems, both pressurized and ambient pressure, are fully automated and allow for push-button catalyst delivery and carbon removal.

pH Matter's CFR system that was developed for on-board air revitalization operates at ambient pressure for safety. The proprietary, transition metal-based catalyst is sprayed onto the walls of the reactor from a central sprayer system as a slurry. The catalyst-coated reactor is then brought to an elevated temperature for catalyst-activation, followed by a carbon growth period of 70 hours. The carbon growth period for one reactor module will produce 1kg of solid carbon, which corresponds to the metabolic conversion of CO_2 for one crew member[4]. Therefore, the 4-CM system developed and delivered to NASA MSFC consists of four reactor modules. An image of this 4-CM system at NASA MSFC can be found in Figure 2 and is described in further detail below.

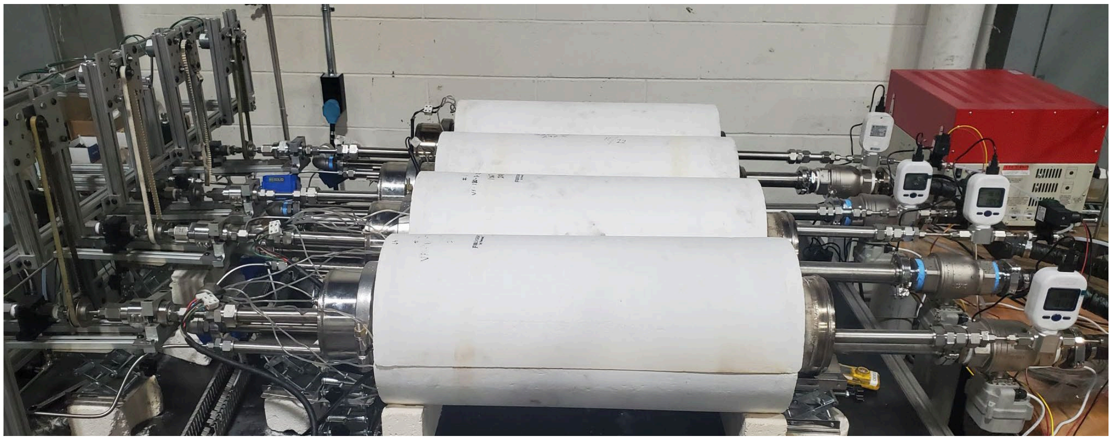

Figure 2. 4-CM CFR System Delivered to MSFC in June 2023

Each CFR has a cylindrical body, 6 inches in diameter and 26 inches long, with a welded-on bottom lid and a removable top lid with a flange and gasket closure system. The reusable graphite composite gasket is stable at the reaction temperatures, and the flange and gasket are held together with a band clamp. Stainless steel tubing extends from the top and bottom lids, comprising gas inlet and outlet ports, carbon removal ports, and the attachment of a rotary union to spin the internal attritor arms. Each CFR has its own custom Thermcraft heating and insulating unit with cast-in heating coils. The Thermcraft units have a vestibule on either end that fits snugly around the body of the reactor. An external control thermocouple is fixed to the outer wall of the reactor for temperature control. One reactor inside its Thermcraft unit can be found in Figure 3.

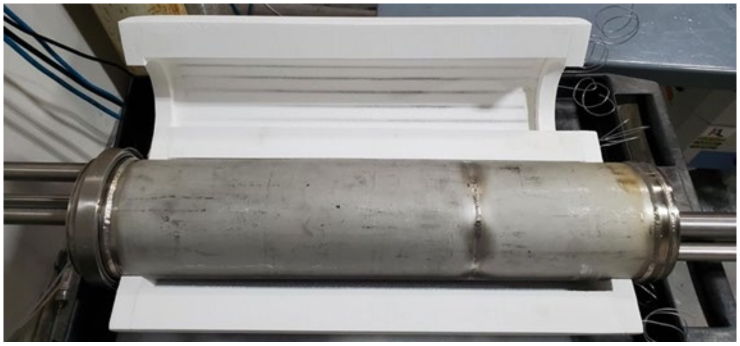

Figure 3. 1-CM Reactor in its Custom Thermcraft Unit[4]

The catalyst delivery system is a key component to the success of the overall CFR system. pH Matter developed a combined attritor arm, catalyst delivery coating attachment . After each catalyst application, the tube is purged with water and then nitrogen is flowed through it for the duration of the carbon growth period. Once the carbon growth period is completed, the process gas is shut off and the reactor cools down under a nitrogen purge. Once the reactor has reached 165ºC and there is no H_2 or CO present in the system, the carbon is removed. This process is completed through a port on the bottom lid of the reactor. A ball valve opens at the port, and the HEPA vacuum removes the carbon while the attritor arms spin to agitate carbon buildup on the walls and funnel carbon towards the port. At the same time, a port in the top lid opens as well to resupply air to the reactor during the vacuum removal process. The outlet gas line is shut during removal to prevent backflow of water from the downstream water trap. The carbon is captured and stored in removable HEPA bags, where it can be used in other processes, moved into long-term storage, or removed and disposed if desired.

One of the major advantages of pH Matter's CFR reactor system is that it can be nearly completely push-button automated. The catalyst delivery and carbon removal processes can occur with very minimal crew member intervention. A crew member still needs to prepare the catalyst slurry and remove and store the accumulated carbon, but every other step can be push-button automated. The controls system consists of P1AM PLC and interfaces with all components in the system using RS-485 communication.

II. Four Crew Member Scale Ambient Pressure CFR System for Onboard Air Revitalization Systems

500-hours of continuous testing of the four-crew member system pictured above was completed at pH Matter before its ultimate delivery to MSFC. This testing consisted of nine carbon removal and catalyst regeneration cycles for each reactor. Each reactor started on a seven hour offset so no more than one reactor was regenerating at any given time. This is important to ensure that adequate oxygen is being produced for the crew. A table showing the regeneration schedule can be found below in Table 1. Catalyst was delivered to each reactor, baked on for one hour, experienced one hour of catalyst activation, and then proceeded to grow carbon for 70 hours. Then, the reactor cooled down for 3 hours under nitrogen and the carbon was removed. After the carbon was removed, the catalyst reapplication process began after a fifteen minute delay. Future improvements will involve optimizing sizing of the catalyst delivery and carbon removal subsystems to minimize crew interaction frequency. Additionally, the run time may be adjusted as more data is generated around carbon growth rate and its effect on system pressure. The nitrogen purge does not need to be pure; it can be pulled from an adjacent recycle stream if available.

Table 1. Regeneration and Carbon Removal Schedule for Four Crew Member Reactor System

Functions	**time (h)**	Stagger time (h)		7		
Cooling Reactor	2		**Reactor 1**	**Reactor 2**	**Reactor 3**	**Reactor 4**
Carbon removal	0.25	Time (hour)	(g Carbon)	(g Carbon)	(g Carbon)	(g Carbon)
Catalyst Coating	0.75	0	0	0	0	0
Catalyst Bake	4	7	322	0	0	0
		14	483	161	0	0
assume:		21	590.3	268.3	107.3	0
46 grams/hr carbon form.		28	670.8	348.8	187.8	80.5
		35	751.3	429.3	268.3	161.0
C removal initiated		42	831.8	509.8	348.8	241.5
		49	912.3	590.3	429.3	322.0
		56	992.8	670.8	509.8	402.5
		63	1073.3	751.3	590.3	483.0
		70	0.0	858.7	697.7	590.3
		77	80.5	939.2	778.2	670.8
		84	161.0	1019.7	858.7	751.3
		91	268.3	0.0	966.0	858.7
		98	348.8	80.5	1046.5	939.2
		105	429.3	161.0	1127.0	1019.7
		112	536.7	268.3	0	1127.0

During the run, it was observed that there was an increase in pressure beyond what was expected. Post-test analysis showed that this was because the outlet tubing was too small at ¼” OD and the flow was being choked. For safety, the flow rate was reduced from a simulated 4-CM flow rate to a 3-CM flow rate. With this new, reduced flow rate, the estimated carbon production for the 4-CM system in 500 hours was 13,200g. The total carbon produced by all four reactors in 500-hours of testing was 12,600g, or 95.5% of the expected value.

Overall, the 500-hour test was successful and identified some opportunities for improvement that the team was able to complete before the delivery of the system to MSFC. First, the outlet gas lines were increased from ¼” to ½”. This will prevent the pressure drop that was experienced during the 500-hour test. The motor timing belt was observed to be weak, stretching out and becoming tangled as the test went on. This was replaced with a chain and sprocket, keeping the same motor. Some of the motorized ball valves seized up during the run, stopping the flow to one of the reactors and preventing changeover to the purge gas on another. This was discovered to be because the valves were wired to be in a constantly energized state, and they eventually overheated. These ball valves were all replaced with self-closing valves that auto-return, which are much less energy intensive and are less likely to overheat and seize. After removing the attritor attachment following the 500-hour test, it was observed that many of the holes were clogged with catalyst. The clogging appeared to start from the bottom up. This makes sense as there was no egress point at the base of the tube. An image of a pristine catalyst delivery attritor attachment can be found below in Figure 4. The center rod has holes drilled along it for the catalyst to be sprayed out. This design will need continued improvement in the future. The 4-CM system was delivered to MSFC in June 2023 to be integrated into their Series Bosch brassboard system. It is anticipated that testing on the overall system will begin in Quarter Three of 2024.

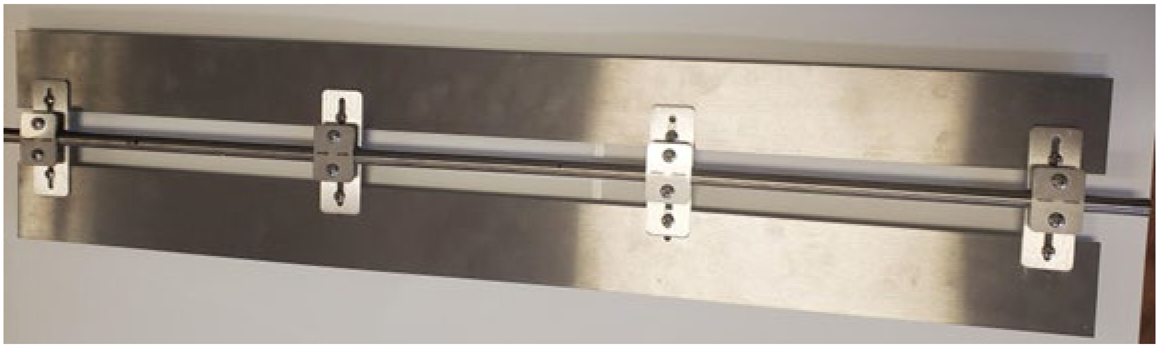

Figure 4. Combined Attritor Arm Catalyst Delivery Assembly

III. Advancements in-Progress for Ambient Pressure CFR System

The next phase of development for the ambient-pressure system is to bring the catalyst delivery and carbon removal subsystems to a more flight-ready status, advancing the TRL of the 4-CM system from TRL 5 to 6. The team also wanted to address the aforementioned clogging of the catalyst delivery tube observed after extended testing. The previous catalyst delivery subsystem consisted of a catalyst reservoir heated on a stir plate that was delivered into the reactor with a diaphragm pump. This subsystem required quite a bit of crew member intervention, mixing the slurry and switching the tube from the catalyst reservoir to the water reservoir to clear the lines. Additionally, diaphragm pumps are not as effective outside of Earth's gravity as there is an internal valve that requires gravity to seat properly. The team has decided to replace the diaphragm pump with a piston pump driven by a pneumatic linear actuator. The pump selected was designed specifically for slurries, has an appropriate draw volume, and can withstand the catalyst temperature at the time of delivery, 45°C. One catalyst delivery subsystem will service all four reactors, and each pre-sealed catalyst reservoir will contain enough catalyst for all four reactors. However, as before only one reactor will be sprayed at a time, maintaining the seven hour stagger.

Dry catalyst and binder powder will be pre-loaded into the bladder bags. A crew member will attach the bladder bag containing enough catalyst for all four reactor regenerations and begin the mixing process. A metered amount of water will enter the bag from the onboard water source and it will be pumped using the piston pump to mix. Once the catalyst, binder, and water have been properly mixed, the crew member will activate the first catalyst delivery spray in the controls system. This will activate the entire program for all four reactors. First, the catalyst will spray into reactor one. All valves will remain closed except for the valves leading from the catalyst reservoir to the piston pump. The linear actuator will draw the pump back to a pre-determined stroke length to load enough catalyst volume for one reactor. The catalyst mixture will be heated inside the pump using an attached heating mat, forming a slurry. The valves leading from the catalyst reservoir are closed and the valve leading to the first reactor is opened. From there, the piston pump will rapidly deploy the catalyst into reactor one. The valve leading to the onboard water supply will open again, and a small amount will be drawn into the piston pump. This water will be sprayed into the reactor to clear the lines. After the catalyst is delivered into reactor one, all valves will close, and the subsystem will be on standby until it is time to deliver catalyst to reactor two. The catalyst reservoir will cool back down to ambient temperature during this seven hour interlude. The catalyst delivery subsystem was approved in a critical design review with MSFC, and fabrication has begun. Once assembled and tested, the subsystem will be delivered and integrated with the 4-CM CFR system at MSFC.

In addition to the catalyst delivery subsystem, pH Matter will make developmental improvements to the carbon removal subsystem. The current method of removing carbon at MSFC is with a mobile HEPA vacuum that is hooked up to each reactor as needed. During carbon removal, a 1" port on the top lid opens to allow air resupply. While this system is effective in removing carbon, there are lingering issues that must be addressed. The first design change that will be implemented will be closing the gas loop during carbon removal. When the carbon is removed from the reactor, the nitrogen that was used as a purge gas is released into the lab through the vents in the HEPA vacuum. While this is fine for on-Earth applications, it will not be ideal for an artificial habitat environment where the relative percentages of nitrogen and oxygen are tightly controlled. Additionally, the purge gas will be lost from the Series Bosch system and will need to be resupplied, adding system mass. The current air resupply strategy is working against process gas conservation as well, as a fifteen minute purge cycle is required to remove oxygen from the CFR system.

Another identified issue that will need a design change is the fact that currently the carbon is difficult to remove from the HEPA vacuum without causing carbon egress, a maintenance task that will be nearly impossible in microgravity using the current subsystem. To solve both problems, the team intends to develop a system consisting of a blower and an atrium of HEPA filters that keeps the purge gas contained within the reactor body and provides an easy way to access the collected carbon. A very elementary schematic of the planned subsystem can be found below in Figure 5. This subsystem will be formally designed and then undergo a critical design review at the end of Quarter Three 2024. The subsystem will then be fabricated, tested, and integrated into the 4-CM CFR system at MSFC with the catalyst delivery subsystem. More robust filters will be added to the outlet gas port as well to prevent dust from interacting with downstream equipment. Parallel work will be completed at pH Matter quantifying the dust released using this new carbon removal strategy, re-engineering if necessary to further reduce dust exposure, and formulating a strategy for long-term on-board storage of the produced carbon.

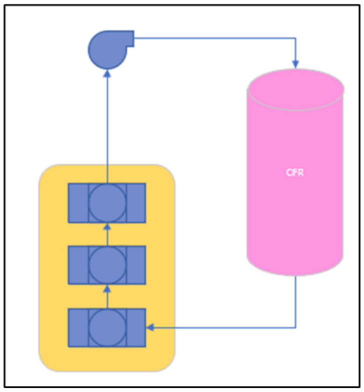

Figure 5. Proposed Closed-Loop Subsystem with Blower and HEPA Filters in an Atrium for Easy Removal

IV. Four Crew Member Scale Pressurized System for In-Situ Resource Utilization

In addition to directly supplying oxygen as part of a Series Bosch system for life support, pH Matter's CFR can be used as part of a reactor series to extract oxygen from Lunar or Martian regolith as part of a carbothermal reduction process[3]. The upstream reactor to the CFR would still be a RWGS reactor, with the carbon dioxide being supplied by either metabolic carbon dioxide or sequestered carbon dioxide from the Martian atmosphere. Reactors will be heated using captured solar energy, focused onto the reactors using a solar concentrator. The carbon produced by pH Matter's CFR will be used as a feedstock for the carbothermal regolith reduction. The water produced could be electrolyzed to release hydrogen and oxygen as well, if desired. The hydrogen would be recycled back to use in the RWGS reactor. This work is being completed with the CUTLAS group at NASA GRC.

For the ISRU application, the 4-CM CFR system was modified to be able to operate at an elevated pressure of 200psia. The reason for this change was that solid carbon production is kinetically favorable at elevated pressures, which would generate more carbon feedstock for the carbothermal reduction process. This was verified in the lab in a 1" reactor, producing approximately twice the amount of carbon per gram catalyst under the same conditions. Additionally, the reactor system will be positioned on the lunar surface rather than in an enclosed environment with humans. This means that pressure is no longer as much of a safety concern, however it is still critical that the system is ASME certified to ensure the safety of the technicians operating it in the lab. The combined catalyst delivery attritor arm system was separated, with the attritors mounted on a solid center rod and modified nozzle extending from the top lid of the reactor.

To achieve the high pressures required for operation, the lid closure system for the four reactors was changed from v-band clamp to bolt-and-nut. The flange, flange bolt size, flange bolt number, flange bolt torque, reactor wall thickness, and reactor welding specifications were chosen based on ASME design code. Each reactor body has the ASME U-stamp. The pressure inside the system was controlled with an automated back-pressure regulator, and the four reactors were operated in series for safety. There is a pressure transducer mounted on each reactor body to monitor the pressure within each reactor, and there are several pressure relief valves within the reactor system that vent to a safe location. The four reactors were positioned in a 2x2 array rather than a 1x4 array for ease of access as well as to reduce the footprint of the overall system. A picture of the 4-CM pressurized CFR system on its test stand can be found below in Figure 6.

Figure 6. Pressurized 4-CM CFR System on its Test Stand in a 2x2 Array Delivered to GRC in February 2024

For this system, although the Thermcraft heating units are installed on the system as before, pictured in Figure 6, the gases will be preheated before entering the reactors. This preheating will occur by concentrating energy on a block with a serpentine channel that the gases will flow through upstream of the reactor. The channels will have copper cladding, so as not to catalyze carbon growth. These blocks will be positioned by the bottom lid of the reactor, one for each reactor body, and then the gases will travel through a tube that is positioned inside the Thermcraft units to the top lid of the reactor, where it will enter. This is so the incoming, heated gases can also make use of heat from the reactor bodies, minimizing heat loss. The Thermcraft units will be used primarily for insulation when the solar concentrator is in operation. However, they can also be used to supplement heat lost to convection during on-Earth testing, a phenomenon that will not be present on the lunar surface. Testing of this system is expected to occur at GRC in mid-2024.

V. ISRU-Scale CFR System

After the fabrication, testing, and delivery of the 4-CM pressurized CFR system, the next step will be to scale up to the ISRU-scale system. ISRU-scale is defined as being able to provide oxygen for forty crew members, using Bosch carbon as the feedstock for carbothermal reduction of regolith, whether lunar or Martian. This corresponds to 0.5kg carbon produced per hour. Additional optimization will be used to determine the tradeoff between reactor weight and the amount of catalyst that would be required for a yearlong mission. In general, smaller reactors require more frequent carbon removal and catalyst regeneration due to the decreased volume. A critical design review of this ISRU-scale system will be completed in March 2024 and fabrication and delivery of the system to GRC is expected to be completed in September 2024.

VI. Next Steps

pH Matter has applied for a research flight mission through NASA TechFlights to test the catalyst delivery subsystem in a relevant environment. The mission would consist of parabolas simulating both lunar and microgravity, which would enable the team to confirm the system works in all environments. The results of these experiments will guide any design changes that may be necessary before the ultimate delivery of the ISRU-scale system to NASA GRC and the catalyst delivery subsystem to MSFC. The team also plans to apply for a second research flight mission in early 2025 to test the carbon removal subsystem and verify that the changes made to the catalyst delivery subsystem were effective.

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