Hydrogen Purification and Recycling for an Integrated Oxygen Recovery System Architecture

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The United States Atmosphere Revitalization life support system on the International Space Station (ISS) performs several services for the crew including oxygen generation, trace contaminant control, carbon dioxide (CO₂) removal, and oxygen recovery. Oxygen recovery is performed using a Sabatier reactor developed by Hamilton Sundstrand, wherein CO₂ is reduced with hydrogen in a catalytic reactor to produce methane and water. The water product is purified in the Water Purification Assembly and recycled to the Oxygen Generation Assembly (OGA) to provide O₂ to the crew. This architecture results in a theoretical maximum oxygen recovery from CO₂ of ~54% due to the loss of reactant hydrogen in Sabatier-produced methane that is currently vented outside of ISS. Plasma Pyrolysis Assembly (PPA) technology, developed by Umpqua Research Company, provides the capability to further close the Atmosphere Revitalization oxygen loop by recovering hydrogen from Sabatier-produced methane. A key aspect of this technology approach is the need to purify the hydrogen from the PPA product stream which includes acetylene, unreacted methane and byproduct water and carbon monoxide. In 2015, four sub-scale hydrogen separation systems were delivered to NASA for evaluation. These included two electrolysis single-cell hydrogen purification cell stacks developed by Sustainable Innovations, LLC, a sorbent-based hydrogen purification unit using microwave power for sorbent regeneration developed by Umpqua Research Company, and a LaNi4.6Sn0.4 metal hydride produced by Hydrogen Consultants, Inc. Here we report the results of these evaluations to-date, discuss potential architecture options, and propose future work.

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Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
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<tbody>
<tr>
<td>AR</td>
<td>Atmosphere Revitalization</td>
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<tr>
<td>C₂H₂</td>
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<tr>
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I. Introduction and Background

Life support is a critical function of any manned space vehicle or habitat. Atmosphere revitalization (AR) technology on the International Space Station (ISS) provides a breathable atmosphere and comfortable living environment for the crew. Prior to 2011, operational functions of the AR system included temperature and humidity control, oxygen generation, trace contaminant control, and carbon dioxide (CO₂) removal. The CO₂ removed from ISS was vented overboard resulting in a loss of 92% of the required respiratory O₂. To resupply this O₂, water was delivered from Earth and electrolyzed in the Oxygen Generation Assembly (OGA). In 2009, a Sabatier CO₂ Reduction system was launched to ISS and became fully operational in June 2011. The Sabatier system interfaces with the OGA and the CO₂ Removal Assembly (CDRA). Carbon dioxide from the CDRA is compressed and stored in tanks until the Sabatier is ready for operation. Hydrogen is produced during the electrolysis of water in the OGA. When the OGA is operational and CO₂ is available in the storage tanks, the Sabatier system is activated and produces methane and water via the Sabatier reaction shown in Eq 1.

Sabatier Reaction

\[ \text{CO}_2 + 4\text{H}_2 \leftrightarrow 2\text{H}_2\text{O} + \text{CH}_4 \]  \hspace{1cm} (1)

The water is condensed, separated, and purified in the Water Processing Assembly and recycled to the OGA to continue producing O₂ for the crew. The CH₄ saturated with water vapor from the condensation process, is vented overboard as a waste product. The addition of the Sabatier reactor to the AR system results in a theoretical recovery of ~54% O₂ from CO₂ and is limited only by the loss of H₂ in the form of vented CH₄ and the loss of uncondensed water vapor in the CH₄ stream. The Sabatier reactor serves to reduce the required water resupply from Earth from ~0.891 kg H₂O/crew member(CM)-day to ~0.459 kg H₂O/CM-day. For long-duration missions beyond Low Earth Orbit, such as Martian transit and surface missions, even more recovery is needed to limit the cost and logistics of resupply. NASA is currently targeting technologies that achieve 75-90% O₂ recovery from metabolic CO₂.¹ One approach to achieve additional recovery is to recycle hydrogen by adding a methane post-processor to the Sabatier-based architecture. NASA has been exploring the Plasma Pyrolysis Assembly (PPA) for this purpose²³. The PPA uses a magnetron to generate an H₂/CH₄ plasma targeting Sabatier CH₄ conversion to hydrogen and acetylene (C₂H₂) as shown in Eq. 2. Secondary reactions with CH₄, as shown in Eqs. 3-5, and reactions with residual water vapor as shown in Eqs 6-7, also occur in the PPA resulting in an effluent mixture containing H₂, unreacted CH₄, product C₂H₂, and trace quantities of H₂O, carbon monoxide (CO), ethylene (C₂H₄), ethane (C₂H₆), and solid carbon (C).

Targeted PPA Reaction

\[ 2\text{CH}_4 \leftrightarrow 3\text{H}_2 + \text{C}_2\text{H}_2 \]  \hspace{1cm} (2)

CH₄ Conversion to Ethane

\[ 2\text{CH}_4 \leftrightarrow \text{H}_2 + \text{C}_2\text{H}_6 \]  \hspace{1cm} (3)

CH₄ Conversion to Ethylene

\[ 2\text{CH}_4 \leftrightarrow 2\text{H}_2 + \text{C}_2\text{H}_4 \]  \hspace{1cm} (4)

CH₄ Conversion to Solid C

\[ \text{CH}_4 \leftrightarrow 2\text{H}_2 + \text{C(s)} \]  \hspace{1cm} (5)

CO Production

\[ \text{C(s)} + \text{H}_2\text{O} \leftrightarrow \text{CO} + \text{H}_2 \]  \hspace{1cm} (6)

CO Production

\[ \text{CH}_4 + \text{H}_2\text{O} \leftrightarrow \text{CO} + 3\text{H}_2 \]  \hspace{1cm} (7)
When the \( \text{H}_2 \) recovered in the PPA product is recycled back to the Sabatier, and the Sabatier is operated at an \( \text{H}_2: \text{CO}_2 \) ratio of 4.25, a theoretical \( \text{O}_2 \) recovery of 91.3% from \( \text{CO}_2 \) is possible. This further reduces the required water resupply to 0.141 kg \( \text{H}_2 \text{O} \)/CM-\text{day}. However, in order to recycle \( \text{H}_2 \) back to the Sabatier, the hydrocarbon byproducts must be removed to prevent fouling of the Sabatier catalyst. In 2015, four sub-scale hydrogen separation systems were delivered to NASA for evaluation for this purpose. These included two electrolytic single-cell hydrogen purification cell stacks developed by Sustainable Innovations, LLC (SI), a sorbent-based hydrogen purification unit using microwave power for sorbent regeneration developed by Umpqua Research Company (Umpqua), and a LaNi4.6Sn0.4 metal hydride produced by Hydrogen Consultants, Inc.

Two of these technology options have been tested at NASA Marshall Space Flight Center and evaluated for relative \( \text{H}_2 \) purification performance, durability, and complexity. Here we report the results of these evaluations, discuss potential architecture options, and propose future work.

II. Hardware and Test Configuration

Four systems were delivered to NASA MSFC for evaluation in an Oxygen Recovery test stand. Two have been tested to-date including the SI Basic Cell Stack and the SI Advanced Cell Stack. A description of the hardware and the testing configuration is provided below.

A. Hardware

1. Sustainable Innovations Electrochemical \( \text{H}_2 \) Separation

The SI cell stacks are both electrochemical \( \text{H}_2 \) separation cell stacks. Electrochemical hydrogen separation provides a means of selectively isolating hydrogen from a mixture of gases. In electrochemical separation, hydrogen is electro-oxidized to protons and electrons, and the resulting protons are electro-reduced in another chamber, combining them with the electrons, thus producing purified hydrogen. The basic technology is well developed, but prior to 2014, was not directly applicable to the PPA product stream due to the relatively significant concentration of carbon monoxide in the product stream. At typical operating temperatures, the carbon monoxide would preferentially adsorb on the catalytic electrodes in the cell, and interfere with their ability to oxidize hydrogen. The carbon monoxide would desorb from the electrodes at temperatures above 150°C, but the acidic polymer that it typically uses as the electrolyte is not serviceable at this temperature. Through a Phase I and Phase III Small Business Innovative Research contract, Sustainable Innovations worked to identify and develop electrolyte materials tolerant to operating at temperatures that are adequate to thermally desorb the carbon monoxide—thus enabling electrochemical hydrogen separation to effectively purify the hydrogen in the PPA product stream. During this effort, two single-cell stacks were developed using unique materials: a Basic Cell Stack, shown in Figure 1, and an Advanced Cell Stack. The materials and design for each are proprietary and will not be discussed in detail here. However, both were covered in Omega heat wrap and insulated using Unifrax (Tonawanda, NY) Fiberfrax S insulation to control stack temperature. The stack was operated using a Sorensen DCS20-60 DC power supply.

B. Test Configurations

1. Stand-Alone Operation

Of the four technology options, only the metal hydride involved stand-alone operations. Other metal hydrides, such as LiH\(^4\) and MgH\(^2\), have been shown to react with acetylene to form metal-carbides under certain conditions. Given the unstable nature of acetylene, and the lack of available literature data on effects of \( \text{C}_2\text{H}_2 \) exposure on the LaNi4.6Sn0.4 metal hydride, a stand-alone safety test was conducted. A test stand was prepared at NASA MSFC’s Explosives Testing Facility. This area was designed for explosives and propellant testing and provided the necessary isolated operation and safety controls to mitigate any violent decomposition event that might occur during testing.
The metal hydride was first charged with H\textsubscript{2} and regenerated to demonstrate nominal thermal and pressure profiles. The metal hydride was then exposed to a gas mixture containing 7\% C\textsubscript{2}H\textsubscript{2}, 1\% CH\textsubscript{4}, and 92\% H\textsubscript{2}. This composition represents a “worst case” C\textsubscript{2}H\textsubscript{2} exposure the metal hydride would see during operation with the PPA. Vessel temperatures and pressures were monitored to compare with the baseline H\textsubscript{2} adsorption data.

2. PPA + H\textsubscript{2} Purification Operation

Each cell stack was integrated with the 2\textsuperscript{nd} Generation PPA for initial evaluation. During this testing, the PPA was operated using compressed ultra-high purity CH\textsubscript{4} and H\textsubscript{2}. The PPA was operated at a 1-CM rate of CH\textsubscript{4} production and an H\textsubscript{2}:CH\textsubscript{4} ratio of 4:1. The reactor was maintained at \~52 torr with an applied microwave power of 550W. This resulted in a PPA effluent containing H\textsubscript{2}, C\textsubscript{2}H\textsubscript{2}, unreacted CH\textsubscript{4}, ethylene, and ethane. Operation in this manner was notably devoid of CO and water vapor. During testing, 100 standard milliliters per minute (SmLPM) slip stream of the PPA effluent was diverted to the stack with flow controlled using a mass flow controller. The resulting H\textsubscript{2} product was evaluated for purity and the process effluent was tested to identify vented composition.

3. SDU + PPA + H\textsubscript{2} Purification Operation

Following PPA + H\textsubscript{2} Purification testing, a Sabatier Development Unit (SDU), designed by Precision Combustion, Inc., was integrated upstream of the PPA. The SDU was operated such that 350 SmLPM CH\textsubscript{4} was produced with no unreacted CO\textsubscript{2}. This resulted in a methane product containing 80 mol\% (dry basis) hydrogen. Water vapor content was maintained at a dew point of 31°C. The PPA was operated identically to PPA + H\textsubscript{2} Purification testing. Because the PPA was fed CH\textsubscript{4} containing water vapor, the resulting effluent contained all previously indicated components. Gas composition data were taken as in the PPA + H\textsubscript{2} Purification testing.

III. Results and Discussion

Two of the four H\textsubscript{2} purification systems were tested at NASA MSFC in 2015 and early 2016. Each technology was evaluated when integrated with the PPA alone, and when integrated with both an SDU and the PPA. The results of this testing and a discussion of its relevance are provided below.

Sustainable Innovations, LLC delivered two cell stacks to NASA MSFC in 2015: the SI Basic Cell Stack and the SI Advanced Cell Stack. A key finding of all testing was that the H\textsubscript{2} purified by the stacks was found to contain no contaminants within the measurable limits of the µGC. This level of purity is highly desirable for recycling to a Sabatier system or PPA with minimal concern for contamination. Differences in performance were observed based on the stack membrane materials themselves, the temperature at which the stack was operated, and the presence of water vapor and CO in the stack inlet gas stream.

A. Basic versus Advanced Cell Stack H\textsubscript{2} Separation Performance

Several data points were taken during operation of each cell stack. These data points varied gas feed from the PPA to the stacks, stack temperature, inlet composition, and applied voltage. For six of the collected data points, the Basic and Advanced cell stacks were held under identical conditions. Percentage of H\textsubscript{2} separation was calculated based on the measured H\textsubscript{2} at the cathode versus the known H\textsubscript{2} feed to the cell stack anode. As can be seen in Figure 2, the Basic cell stack demonstrated a greater percentage of H\textsubscript{2} separation than the Advanced cell stack in all cases. This data correlates well with that reported by the cell stack vendor, Sustainable Innovations, LLC where the Basic

**Figure 2.** H\textsubscript{2} separation performance comparison between Basic and Advanced cell stack.
stack demonstrated better H\textsubscript{2} separation performance than the Advanced stack. SI attributed this observed difference in performance to the relative effectiveness of the membrane material used in each cell stack.

B. Process Effluent Composition

In addition to measuring H\textsubscript{2} separation across each stack, gas composition data was taken for the anode inlet (feed from the PPA), anode outlet (H\textsubscript{2} lean) and the cathode outlet (separated H\textsubscript{2} product). As mentioned above, the separated H\textsubscript{2} product was pure beyond the evaluation means of the GC. The anode inlet composition varied for several reasons. First, when the PPA operates, the outlet composition changes slightly over time due to byproduct carbon formation in the PPA reactor. High reflected power with in the PPA reactor is an indicator of excessive carbon build-up. When this occurs, the PPA is regenerated. However, several hours of operation occur between regenerations. This provides several hours over which the PPA outlet composition changes. The PPA outlet composition will also vary based on integration with the Sabatier. When Sabatier effluent is fed to the PPA, the PPA effluent can vary further due to the presence of water vapor and CO in the gas feed. Average PPA effluent compositions are shown in Figure 3. As can be seen from the graph, the presence of water and CO in the PPA feed results in high CO concentration and inhibits C\textsubscript{2}H\textsubscript{2} production from CH\textsubscript{4}.

Figure 3. Stack inlet (PPA effluent) composition as a function of level of integration.

Given these variations in inlet composition, the effluent from the anode would be expected to mirror the relative compositions of the gases with the exception of H\textsubscript{2}. However, during testing of both the Basic and Advanced cell stacks, the outlet composition from the anode was considerably different than anticipated. In all cases, high levels of C\textsubscript{2}H\textsubscript{4} and C\textsubscript{2}H\textsubscript{6} were observed with minimal or no C\textsubscript{2}H\textsubscript{2}. The overall chemistry of formation of C\textsubscript{2}H\textsubscript{4} and C\textsubscript{2}H\textsubscript{6} from CH\textsubscript{4} is shown in equations 3 and 4 above. However, the actual process of producing these hydrocarbons involves free radical intermediates. As shown in Eq. 8, CH\textsubscript{4} forms CH\textsubscript{3}* free radicals which then recombine to form C\textsubscript{2}H\textsubscript{6}. Similarly, C\textsubscript{2}H\textsubscript{4} is converted to C\textsubscript{2}H\textsubscript{6} and C\textsubscript{2}H\textsubscript{4} is converted to C\textsubscript{2}H\textsubscript{2}. The reverse reactions also occur providing a mechanism for hydrogenation of C\textsubscript{2}H\textsubscript{2} to the other hydrocarbons.

\text{Ethane Formation from CH}_4 \quad \text{CH}_4 + \text{CH}_4 \leftrightarrow \text{CH}_3^* + \text{CH}_3^* + \text{H}^* + \text{H}^* \leftrightarrow \text{C}_2\text{H}_6 + \text{H}_2 \quad (8)

While hydrogenation of C\textsubscript{2}H\textsubscript{2} does not pose any safety concerns, it is highly detrimental to a proposed Sabatier + Plasma Pyrolysis Assembly architecture. Conversion of metabolic CO\textsubscript{2} to C\textsubscript{2}H\textsubscript{2} results in a theoretical maximum of \textasciidot86% O\textsubscript{2} recovery from CO\textsubscript{2}, conversion of CO\textsubscript{2} to C\textsubscript{2}H\textsubscript{4} results in a theoretical maximum of only \textasciidot72%, and conversion to C\textsubscript{2}H\textsubscript{6} results in a theoretical maximum of only \textasciidot61.5%. In order to meet NASA’s mission targets, a minimum of 75% O\textsubscript{2} recovery is required. Conversion to C\textsubscript{2}H\textsubscript{4} or C\textsubscript{2}H\textsubscript{6} will not meet this target, making a Sabatier + PPA architecture inadequate for future long-duration manned missions. Thus, in order to be considered for the architecture, the cell stacks must not hydrogenate C\textsubscript{2}H\textsubscript{2}.

Hydrogenation products from C\textsubscript{2}H\textsubscript{2} were observed in the effluent of both cells. Three theories were proposed as possible causes of the observed phenomenon in the stacks. First, in previous testing, the cell stacks were shown to produce byproducts at lower temperatures. If this were the case, raising the temperature of the stack could decrease the hydrogenation reactions. Second, it was possible that excess H\textsubscript{2} was available during separation. Insufficient voltage to move the H\textsubscript{2} across the membrane would result in a net H\textsubscript{2} surplus at the surface of the membrane. This localized H\textsubscript{2} combined with the voltage applied to the cell could have been contributing to the hydrogenation of
Finally, platinum was used for electrodes within the cell stack. Platinum has been shown to catalyze the hydrogenation of C$_2$H$_2$ under vastly different circumstances, but the presence of the metal under the conditions may have contributed to the observed results.

Testing was first conducted to determine the effect of stack temperature on the relative hydrogenation of C$_2$H$_2$. Figure 4 shows the results of data points collected on the Advanced cell stack. All conditions for each data point were identical with the exception of temperature. As can be seen, the higher temperatures resulted in both more hydrogenation of the C$_2$H$_2$ and of the production of more hydrogen-rich molecules (e.g. CH$_4$). Operation of the stacks at temperatures lower than 160°C might result in a marked decrease in hydrogenation effects. However, the high temperature is critical for the H$_2$ separation technology due to the CO in the effluent stream. At lower temperatures, CO poisons the membrane and H$_2$ separation is compromised. This makes operation at a lower temperature unfeasible for a long-term solution.

Testing was then conducted to explore the effect of overvoltage on hydrogenation based on the theory that excess H$_2$ at the surface of the membrane was contributing to the conversion of the C$_2$H$_2$. For this test, the total flow of PPA effluent to the cell stack anode was limited to very low flow rates to achieve near 100% H$_2$ separation (69 SmLPM, 60 SmLPM, and 48 SmLPM) and the voltage varied (0.00V, 0.16V, 0.30V, 0.46V, and 0.60V). The results show an interesting phenomenon. First, regardless of the flow rate and the voltage tested, all of the C$_2$H$_2$ was hydrogenated to some degree, resulting in no measurable C$_2$H$_2$ in the stack outlet stream. Second, regardless of the flow rate and the voltage tested, the total quantity of C$_2$H$_2$ converted to CH$_4$ did not change, as seen in Figure 5. The voltage did, however, appear to have an effect on the degree to which C$_2$H$_2$ was converted to C$_3$H$_4$ and C$_2$H$_6$ as can be seen in Figure 6 and Figure 7, respectively, but was independent of flow rate. An increase in voltage reduced the total hydrogenation as evidenced by the increase in C$_3$H$_4$ and relative decrease in C$_2$H$_6$ as voltage increased. While this would be a promising result if the goal were to simply decrease
hydrogenation, the fact that all of the C\textsubscript{2}H\textsubscript{2} is still being converted in the process implies that an overvoltage alone will not eliminate the problem.

A third possibility is that the Pt catalyst in the stack is causing the observed hydrogenation. If this were the case, two options might be taken to reduce or eliminate the problem. First, the catalyst material could be changed. Unfortunately, other catalysts known to be H+ conductors including Palladium and Silver are not viable options for this application. Palladium, like Pt, is a known hydrogenation catalyst. Silver, on the other hand, is known to react violently with C\textsubscript{2}H\textsubscript{2}. The second option would be to limit the mass transport of C\textsubscript{2}H\textsubscript{2} to the catalyst surface. This might be accomplished through the use of a gas diffusion layer of greater thickness. The thicker layer would limit mass transport of the larger molecules (e.g. C\textsubscript{2}H\textsubscript{2}) to the surface, while allowing the smaller molecules (e.g. H\textsubscript{2}) ready access to the catalyst surface. In an effort to further understand the phenomenon, the Basic Cell Stack was returned to SI for evaluation and refurbishment with a thicker gas diffusion layer.

Finally, during the course of testing, it was discovered that the presence of H\textsubscript{2}O and/or CO in the gas stream limited hydrogenation of C\textsubscript{2}H\textsubscript{2}. This can be seen in Figure 8, where residual C\textsubscript{2}H\textsubscript{2} was observed when water and CO were present in the feed stream. While it is not yet clear what is causing this phenomenon, it is possible that CO, known to adsorb to the catalyst surface, blocks reaction sites that would otherwise be used to hydrogenate C\textsubscript{2}H\textsubscript{2}. If this is the case, one could assume that H\textsubscript{2} separation would also decrease with the reduced reaction sites. This is, in fact, what was observed. If water vapor plays a role in this phenomenon, it is unlikely that water is electrolyzed at the catalyst surface given the low voltages. However, protons are known to be generated at the anode, and if water is present, hydronium ions (H\textsubscript{3}O\textsuperscript{+}) are likely formed and surrounded by other water molecules. In this scenario, there is no driving force to reduce C\textsubscript{2}H\textsubscript{2} or any other species. This may help explain the relatively lower C\textsubscript{2}H\textsubscript{2} hydrogenation.
IV. Architectural Options

Three primary options for hydrogen separation technology to be used in conjunction with the PPA are being considered. These options include the Sustainable Innovations electrochemical cell stacks, the Umpqua sorbent based H₂ separation system, and a system based on metal hydrides. The system architectural requirements for these three systems are discussed below.

A. Sustainable Innovations Basic and Advanced Cell Stacks

Figure 9 shows the general system architecture required to incorporate an electrochemical hydrogen separation cell stack into the PPA system architecture. This is by far the simplest system architecture since the cell stack would operate continuously, in contrast to the batch-processing necessary for a sorbent or metal hydride based system.

Continuous operation would eliminate the valves necessary for cycling the sorbent or metal hydride beds. Also, since the cell stack would be capable of sourcing hydrogen at pressures well above the minimum required by the CRA and PPA no compressor is necessary in the system. The cell stack membranes are compatible with the water vapor in the PPA effluent stream, and in fact require a trace amount of water vapor for proper operation, so would...
not require a desiccant stage. The use of which would, of course, mean that some water vapor would be lost to space.

**B. Umpqua Sorbent-Based H₂ Purification**

Figure 10 shows the general architecture required for a sorbent-based hydrogen separation architecture. A set of two sorbent beds, one adsorbing and one thermal/vacuum desorbing, would be required for continuous operation. The Umpqua concept uses microwaves instead of conventional heaters to speed heating of the sorbents during regeneration. Zeolite sorbents would capture acetylene and allow hydrogen and any residual methane to pass into the recycle loop. The passage of residual methane is actually beneficial since it would allow for the re-processing of any methane that was not converted to acetylene during the first pass through the PPA. A sorbent-based architecture would require a pressure controller to maintain pressure in the sorbent bed during operation, an accumulator tank to act as a buffer volume to balance process flows, and a compressor to provide sufficient pressure in the sorbent beds for adequate adsorption.

![Sorbent-based H₂ separation architecture.](image)

**Figure 10. Sorbent-based H₂ separation architecture.**

Additionally, since zeolite sorbents have a high affinity for water vapor, a desiccant stage would be needed upstream of the zeolite beds. This desiccant stage could be integrated into the inlet side of the zeolite beds, which would add minimal complexity to the system architecture but would cause a net loss of water vapor since it would be desorbed to space vacuum when the beds were regenerated. The desiccant stage could also be designed as separate beds, which would add system and operational complexity but would allow for water vapor to be returned to the cabin during regeneration.

It should also be considered that a dual swing sorbent bed system has considerable valving challenges, which are not shown schematically in Figure 10. Manifolds and valves must be provided to effectively isolate and cycle the beds between operation and regeneration modes. The dust produced by swing-bed sorbent systems can be a significant operational issue for valves as can be seen in the operational history of the Carbon Dioxide Removal Assembly.

**C. Metal Hydride H₂ Purification and Storage**

Figure 11 shows the general architecture required for a metal hydride based hydrogen purification architecture. In a metal hydride architecture PPA effluent would flow to a metal hydride bed that would capture hydrogen and allow all other constituents to flow out to space vacuum. A second bed would be heated to supply hydrogen to the hydrogen recycle loop. A third bed would be exposed to space vacuum at elevated temperatures to condition the bed for further hydrogen processing. A buffer volume would still likely be necessary in this architecture but the volume could be smaller and lower pressure than the volume needed in the sorbent-based architecture. No compressor would be needed since the thermal desorption of hydrogen from a metal hydride can supply hydrogen at the pressure necessary for the operation of the PPA and CRA. Valve and manifold complexity would be similar to the sorbent system, however, it is unlikely that there would be dusting issues from the metal hydrides. Heating and cooling the metal hydrides would be less challenging than sorbents due to their higher thermal conductivity and lower volume.

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The presence of water vapor present in the PPA effluent stream must still be addressed in metal hydride architecture. Water-tolerant metal hydrides may be available and would simplify the system architecture but would allow water vapor loss to space vacuum. A separate desiccant system could be incorporated up-stream of the metal hydride system, which would allow water vapor to be returned to the cabin albeit at the cost of power and system complexity.

V. Future Work

Four technologies were identified as potential solutions for H₂ separation and recycling in a Sabatier post-processing O₂ recovery architecture. Two of these technologies have been tested. Future work will include a comparable evaluation of the Umpqua H₂ Separation system and the metal hydride. Initial findings of the Sustainable Innovations, LLC cell stacks show the capability to separate nearly 100% of the H₂ fed to the stack and extremely high H₂ product purity. However, the stacks also demonstrate hydrogenation of PPA product C₂H₂ yielding a lower architectural O₂ recovery efficiency. Ongoing efforts will seek to identify methods to reduce or eliminate the hydrogenation of C₂H₂ while maintaining H₂ separation and product purity.

Acknowledgments

The authors would like to acknowledge Kenny Bodkins, Tom Williams, and Jeff Richardson for their technical support in maintaining the test facility and software. This work was funded with Small Business Innovative Research contracts and the Human Exploration and Operations Mission Directorate’s Advanced Exploration Systems Program’s Life Support Systems Project.

References


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