Development of a Microwave Regenerative Sorbent-based Hydrogen Purifier

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This paper describes the design and fabrication of a Microwave Regenerative Sorbent-based Hydrogen Purifier (MRSHP). This unique microwave powered technology was developed for the purification of a hydrogen stream produced by the Plasma Pyrolysis Assembly (PPA). The PPA is a hydrogen recovery (from methane) post processor for NASA's Sabatier-based carbon dioxide reduction process. Embodied in the Carbon dioxide Reduction Assembly (CRA), currently aboard the International Space Station (ISS), the Sabatier reaction employs hydrogen to catalytically recover oxygen, in the form of water, from respiratory carbon dioxide produced by the crew. This same approach is base-lined for future service in the Air Revitalization system on extended missions into deep space where resupply is not practical. Accordingly, manned exploration to Mars may only become feasible with further closure of the air loop as afforded by the greater hydrogen recovery permitted by the PPA with subsequent hydrogen purification. By utilizing the well-known high sorbate loading capacity of molecular sieve 13x, coupled with microwave dielectric heating phenomenon, MRSHP technology is employed as a regenerative filter for a contaminated hydrogen gas stream. By design, freshly regenerated molecular sieve 13x contained in the MRSHP will remove contaminants from the effluent of a 1-CM scale PPA for several hours prior to breakthrough. By reversing flow and pulling a relative vacuum the MRSHP prototype then uses 2.45 GHz microwave power, applied through a novel coaxial antenna array, to rapidly heat the sorbent bed and drive off the contaminants in a short duration vacuum/thermal contaminant desorption step. Finally, following rapid cooling via room temperature cold plates, the MRSHP is again ready to serve as a hydrogen filter.

Nomenclature

AC	=	Activated Carbon
ºC	=	Degrees Celsius
CH₄	=	Methane
cm	=	Centimeter
CM scale	=	Crew Member scale (In current context, a 1-CM scale PPA operates at a 350 sccm methane pyrolysis rate with a 4:1 hydrogen to methane feed ratio; higher PPA CM scales have proportionally high methane rates, but same gas feed ratio.)
CRA	=	Carbon dioxide Reduction Assembly
g	=	Gram
H₂	=	Hydrogen
L	=	Liter
MFC	=	Mass Flow Controller

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mL	=	Milliliter
mol sieve	=	Molecular Sieve
MSFC	=	Marshall Space Flight Center
NASA	=	National Aeronautics and Space Administration
OGA	=	Oxygen Generation Assembly
PPA	=	Plasma Pyrolysis Assembly
QMS	=	Quadrupole Mass Spectrometer
sccm	=	Standard Cubic Centimeters Per Minute
SBIR	=	Small Business Inovative Research

I. Introduction

T HIS paper summarizes the results of a post SBIR Phase 1 development effort to purify hydrogen produced by the Plasma Pyrolysis Assembly (PPA). In the Phase 1 a selection of potential sorbent materials were screened for their effectiveness for purifying hydrogen from an ersatz gas mixture representative of the PPA effluent. The sorbent materials evaluated included several molecular sieves (mol sieves), activated carbons and high surface area forms of silica and alumina. Of the sorbents tested, molecular sieve 13X was the most desirable option [1].

Sorption media was to be regenerated by microwave heating under vacuum. Implementation of microwave desorption techniques was to be developed under a follow-on Phase 3 sub-scale development effort. The use of microwave power for bed regeneration offers several advantages over conventional thermal regeneration using resistive heating elements. The benefits of utilizing microwaves include: 1) The entire volume of the bed is heated in unison. 2) Heat is distributed more uniformly. 3) The sorption media is selectively heated so little microwave power is lost to the surroundings. In conclusion, H_2 purification coupled with microwave dielectric heating for efficient thermal desorption of the contaminants from the media is an attractive approach to meeting NASA's needs.

Development of a sub-scale device to demonstrate Microwave Regenerative Sorbent-based Hydrogen Purifier (MRSHP) technology was performed. The final device is shown in Figure 1. By utilizing the well-known high sorbate loading capability of conventional physical sorbents coupled with microwave dielectric heating phenomena, MRSHP technology is employed as a regenerable filter for a contaminated hydrogen gas stream. The design, fabrication and delivery to NASA, of a sub-scale MRSHP device in the time

Figure 1. MRSHP deliverable prototype.

interval intermediate to the Phase 1 and Phase 2 efforts was conducted to support an aggressive development schedule which targeted delivery of a full 4-CM scale MRSHP prototype by the end of the Phase 2 project. Concurrent testing performed early in the Phase 2 at NASA-MSFC using the sub-scale MRSHP device attached to a similarly sized Plasma Pyrolysis Assembly (PPA) was to provide important research and design guidance during the Phase 2 project and help assure a successful outcome.

In order to prepare for NASA's projected manned Mars mission slated for mid-2030s, several Air Revitalization (AR) system elements must be advanced to fully flight certified status. According to mission planners key decision points for hardware will occur in the 2017-2018 timeframe. Included in this list of AR elements is the PPA and associated hardware, with the hydrogen purification assembly deemed of paramount importance. In order to achieve this critical objective in the time remaining, several key hardware demonstration tests are required, including 3 years of operational time aboard ISS. To support this schedule, a 4-CM scale hydrogen purification device capable of integration testing with the advanced 3rd generation PPA [2] is required by the end of fiscal year 2017 (September).

II. Background

The Sabatier reaction is used for the recovery of water from carbon dioxide, by reduction with hydrogen. During the process, half of the hydrogen is lost as methane. To recover this valuable resource, the PPA was developed to extract H_2 from CH_4 produced by the Carbon dioxide Reduction Assembly (CRA). The PPA was designed to selectively generate gas-phase products, primarily hydrogen and acetylene with minor quantities of ethylene, unreacted methane, and other hydrocarbons. The work presented in this paper was directed toward purifying this H_2 effluent with the use of regenerable sorption media. Once purified, some of the H_2 is returned to the PPA and the remainder passed to the CRA to augment H_2 supplied from water electrolysis which takes place in the Oxygen Generation Assembly (OGA).

Utilization of physical sorbents for contaminant removal from PPA effluent is not a new concept, having been investigated by NASA in recent years [3]. Similarly, the application of dielectric heating to induce thermal desorption of contaminants has been explored by the lead author in previous work [4]. However, the use of microwave dielectric heating to regenerate PPA effluent filter media has only recently been considered [5]. During steady-state operation the PPA recovers up to 75% of the hydrogen contained in the methane byproduct

$$CH_4 \rightarrow 1/2 \ C_2H_2 + 3/2 \ H_2 \ , \tag{1}$$

that results from Sabatier reduction of metabolic CO_2 to water

$$CO_2 + 4 \ H_2 \rightarrow 2 \ H_2O + CH_4 \ , \tag{2}$$

during the primary oxygen recovery step in an advanced air revitalization process [6]. As natural products of PPA operation, several gaseous compounds are created along with the liberated hydrogen in the reactive plasma environment. These compounds include various hydrocarbons, most notably: acetylene (via the principal reaction Eq. (1)), unconverted methane, ethylene and ethane. In addition, due to trace amounts of water vapor and CO_2 contained in the PPA influent, as part of the methane stream coming from the upstream Sabatier, water vapor and carbon monoxide (formed from CO_2 in the plasma) are present in the PPA effluent [7]. Since relatively pure recycled hydrogen is required as an input for both the Sabatier and PPA, a means to remove this contaminant load is required. Both activated carbons and molecular sieves have demonstrated significant sorption capacities for these compounds, with carbon monoxide being the notable exception. The presence of low levels of carbon monoxide recycled to the Sabatier, however, is not a problem since the Sabatier reduction reactor will effectively reduce it, along with the primary carbon dioxide reactant, to water and methane.

MRSHP technology is a candidate hydrogen purification approach that has received recent interest from NASA's AR community in the form of a SBIR Phase 1 proof of concept project that concluded in December of 2014. By building off the foundation of this fruitful 6 month project and our experience maturing novel microwave technologies [8-27], the sub-scale development effort described herein was intended to bolster the likelihood of successfully developing a fully functional 4-CM scale hydrogen purification device by the end of the Phase 2 effort (mid-2017). Historically, due to the limited 2-year development timeframe of a typical SBIR Phase 2 project, only a subscale (e.g., <1-CM) prototype is provided as a final deliverable. To achieve a higher capacity device in this time period is often difficult due to the novel, unproven nature of the proposed technology. This inherent limitation follows directly from the nature of the SBIR program itself, whose focus is to promote innovative solutions to engineering problems and therefore often literally requires building a technology from the ground up. Our Phase I

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effort, however, has successfully demonstrated both effective contaminant removal and the regenerative nature of candidate sorption media at a level (0.1-CM scale) more typical of a Phase 2 effort than a Phase 1. As such, by building off this moderate success, the goal of assembling a 1-CM scale device during a 6 month gap project was deemed feasible.

III. Results of Phase 1 investigation

In the Phase 1 work six activated carbons were evaluated, two spherical and four granular. The activated carbons were produced under different conditions and thus displayed different sorption properties. Four molecular sieves were evaluated, 3A, 4A, 5A and 13X, all obtained from Grace Davison. High surface area forms of alumina and silica were evaluated, BASF CPN 14x28 alumina and Fuji Silycia CARiACT Q-30C silica. Silica gel from Grace Davison was used for water removal. Breakthrough curves were generated by passing gas mixtures through 111 cm^3 sorbent beds until the compounds in the influent gas mix appeared in the effluent. The breakthrough curves were used to determine the sorption capacities of each sorbate on each sorbent at several temperatures. The sorbates that were evaluated for the hydrogen purification portion of this project were acetylene, methane, ethylene and carbon monoxide.

The best sorbent to be used for hydrogen purification was found to be molecular sieve 13X. This material displays 4 times the capacity of molecular sieve 4A, the sorbent previously considered for acetylene removal from hydrogen. The most probable implementation resulting from the Phase 1 research was thought to be the use of silica gel to remove water vapor and molecular sieve 13X to remove acetylene, ethylene and related hydrocarbons, while allowing hydrogen and methane to pass through. Thermal regeneration of silica gel and mol sieve 13X, loaded with target sorbate contaminants, was demonstrated as was microwave heating of sorbent material. The longevity of the mol sieve 13X media was subsequently demonstrated with repeated cycles of sorption and desorption. These exciting results demonstrated the practicality of sorbent based hydrogen purification coupled with microwave desorption of the sorbent media.

IV. Sub-Scale Prototype Development

Because of the rapid development cycle necessitated by delivery of prototype hardware at the end of a six month project, every step of the effort required expeditious completion and rigid adherence to schedule. Even though the prior 6 month Phase 1 work identified 13x media as the optimal sorbent with a significant microwave susceptibility, many of the important features known to be required in a successful design were either completely untested or only marginally so. These included i) determination of the need to mix microwave susceptible activated carbon with the 13x to attain sufficient heat up; ii) the use/design/quantity/placement of antennae within the media; iii) the balanced distribution and transference of power from a waveguide source through multiple cables to yet another waveguide based sorbent bed with varying impedance loads; iv) the insertion of thermocouple into the sorbent media contained in a waveguide; v) the manner in which the media could be quickly returned to room temperature and made ready for continued hydrogen purification; and vi) the sizing of the bed combined with the placement of the all these items in order to effect efficient, uniform heating of the media with a maximum regenerable sorbent volume and rapid post regeneration cool-down. Long lead time items required from suppliers (some at 10-12 weeks) had to be identified early in the design phase and ordered once we were sufficiently confident that they would be necessary in the final design, which was difficult because so many of the unknowns listed above, if not favorably resolved, could drastically impact the final design. Risk mitigation was greatly aided by our ability to computer model multiple design aspects. The ability to perform modeling in parallel with process design was crucial to success of this short-fuse project. Models were prepared in Comsol Multiphysics using the RF physics module and evaluated to glean information ranging from microwave power transference to the media through coaxial cables and antennae, to the complex multiphysics coupled interactions between electromagnetic and thermal phenomena within the media. Where possible, empirical testing was used to confirm operation predicted by models. Nevertheless, even with all these factors coming together to aid this aggressive development schedule, there were many operational elements whose functionality was not truly evaluated until final prototype testing near the end of the contact. Fortunately, these unknowns worked as intended and very few design alterations/concessions were required prior to prototype delivery. In fact, with the notable exception of finding/defining the power and temperature limits for the coaxial cables, and a last minute anomaly that resulted from a refurbishment of the sorbent bed with an inappropriate Teflon dielectic just prior to crating the prototype, final assembly and checkout of the device occurred relatively seamlessly requiring only a two week extension of the contract.

An important determination, that was required up front in the design work was the whether or not the 13x media alone would heat up sufficiently by direct microwave application at moderate power levels. If not, then a

microwave susceptible material would need to be added to the sobent media to bring the 13x to temperatures required for adequate vacuum regeneration (T > 200 °C). In the Phase 1 work, activated carbons (known microwave susceptors) were found to also have a significant sorption capacity for the contaminants in the PPA hydrogen effluent. As such, while significantly less than the 13x, the addition of an AC would otherwise be an acceptable compromise. This media dilution/mixing was not required however. Experiments performed using small aliquots of virgin 13x media taken directly from the supplied canisters were tested in a microwave oven. Heat up profiles for these experiments are the yellow and pink curves shown in Figure 2. Clearly, for both Grace Davison and Delta media, a maximum temperature is reached within a few minutes, that remains below 400 ºC. At the time it was unclear whether this effect was due to cooling of the media during successive temperature measurements or truly a dielectric temperature dependency was unclear. For further clarification, a second set of experiments was carried out that again varied the 50% power microwave exposure times but, rather than taking successive temperature measurements, started with room temperature media each time and only acquired a temperature reading at the end of each uninterrupted full exposure time interval. These data (purple and brown curves in Figure 2) showed a linear time vs. temperature response, for temperatures to over 400 C. Indicating that over most of the temperature range of interest (T<550 C; which is the purported calcination temperature used in the preparation of Grace Davison 13x) the microwave susceptibility does not decrease with increasing temperature. Most importantly, dilution with AC is not needed for our application.
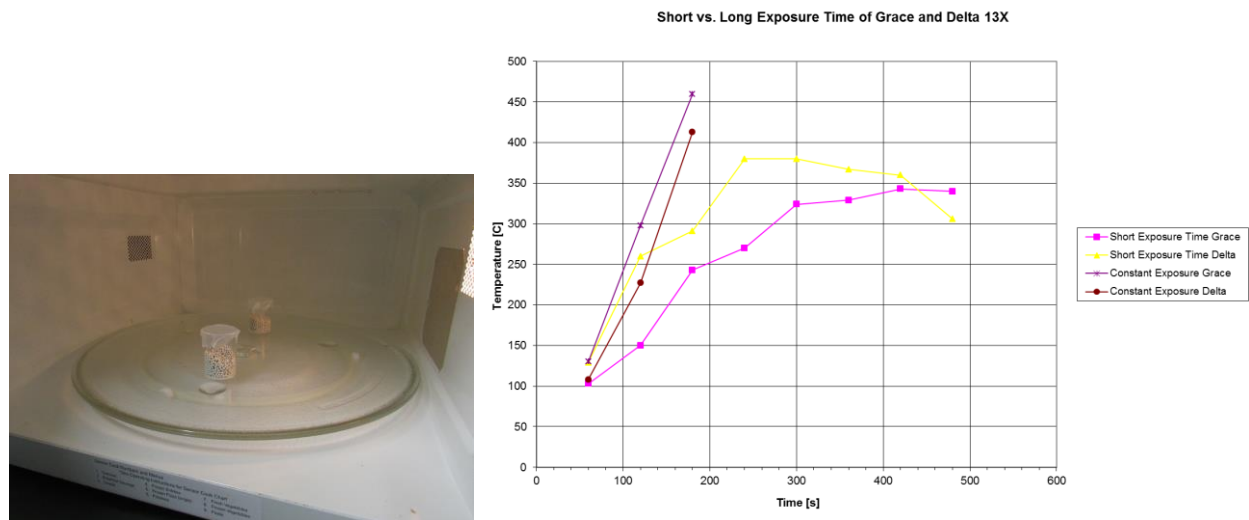
Figure 2. Microwave Heating of both Grace Davison and Delta 13x Media as Function of Exposure Time. Testing using Microwave Oven at 50% Power (Upper Photos) and Resulting Temperature Curves as a Function of Time (Lower Plots).

The same test section used in the dielectric properties evaluation was further modified to allow for temperature profile measurements acquired after microwave heating. Photos of this test section and the test apparatus used to acquire the temperature profile down the length of the test section are shown at the top of Figure 3. By gradually inserting the thermocouple probes directly after removing microwave power, temperature variation both down the length of the test section as well as across its width (the broad-wall dimension) was recorded. A computer model of this apparatus was also prepared and evaluated as depicted by the images located in the center of Figure 3. The results from this simulation were compared against the measured temperature profiles acquired in the lab, as shown by the plots at the bottom of Figure 3. While not yielding exact matches, the computer model does predict temperatures within 15% of measured values as well as similar phenomena to what was observed in the lab. In each case, the cooling effects of the quartz windows in direct contact with the media at both ends of the test section is supported by the lower temperatures at the front end of the media. Similarly, the oscillatory nature of the observed temperature profile is also seen in the model results. The source of this variation is apparently due to the reflected power that occurs because of the quartz window located at the back end of the test section. Reflected and incident waves superimpose to give a standing wave with its consequential hotter/cooler heating pattern.
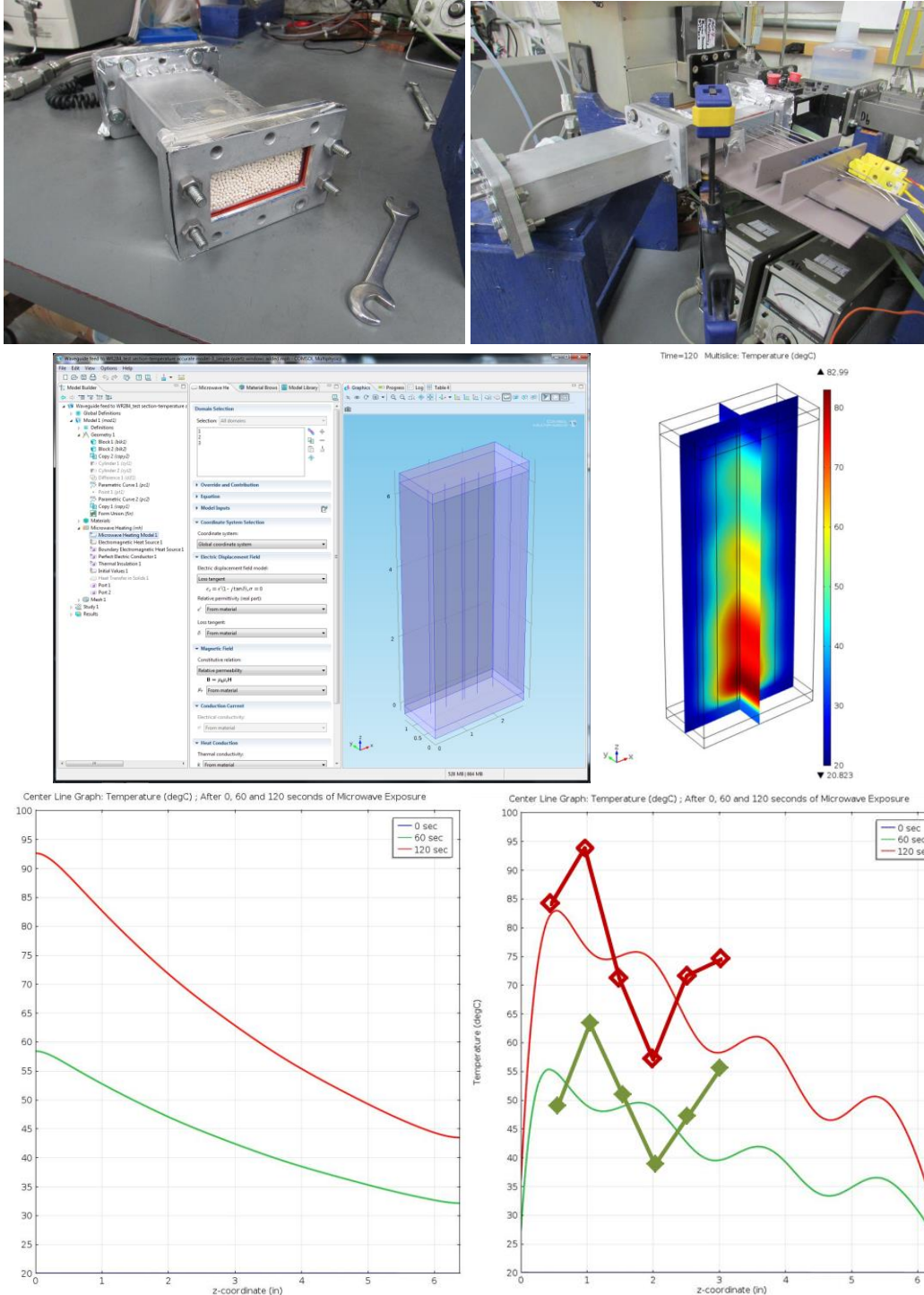
Figure 3. Temperature distribution profiles resulting from microwave heating. <u>Top Photos</u>: (Left-) 13x media filled WR284 test section and (Right-) microwave heating test stand. <u>Middle Images</u>: (Left-) Comsol model of test section and (Right-) resulting temperature profile. <u>Lower Images</u>: Results (Left-) without and (Right-) with quartz windows; model predictions (no symbols) and lab measurements (w/ symbols), each measured along centerline of waveguide.

In the Phase 1 work 1/16 inch thermocouple probes were inserted into our waveguide test beds to measure the temperature profile <u>during</u> microwave heating. On occasion it was observed that the probes closest to the microwave source would overheat or otherwise be adversely affected by the EM field, presumably due to

insufficient wall thickness shielding the thermocouple wires contained within. Subsequently, in following tests, these probes were inserted only after removal of microwave power. For the prototype however, these probes need to be left in position even during microwave desorption. To assess the feasibility of using thermocouple probes in a media filled waveguide, the computer model prepared for dielectric testing was modified to include 1/8 inch diameter thermocouples inserted at various positions. It was found that when inserting the thermocouples through the narrow wall of the waveguide, there was minimal impact on power transfer and the resultant thermal profile within the media. In contrast, when the probes were inserted through the broad wall of the waveguide, the power and temperature profiles varied greatly from the control case containing no probes. Figure 4 contains temperature profiles for comparisons between each of these cases. While probes introduced through the narrow-wall have minimal impact, those inserted through the waveguide broad-wall produce temperature extremes of over 1,000 ºC localized around the probes closest to the microwave inlet. This is not terribly surprising since this same configuration is used in the operation of the PPA to help trap plasma formation to the end of a metal stub. At the root of this phenomena is the orientation of the electric field that is established for the TE10 mode, which is parallel with a stub inserted through the broad-wall and therefore acts as a short-circuit to the EM wave energy. This investigation showed that we can safely use permanently located metal probes in the waveguide as long as they are well grounded with the waveguide and inserted in an orientation that is perpendicular to the electric field. As such, 1/8 inch thermocouple probes of sufficient wall thickness were used in the final design.
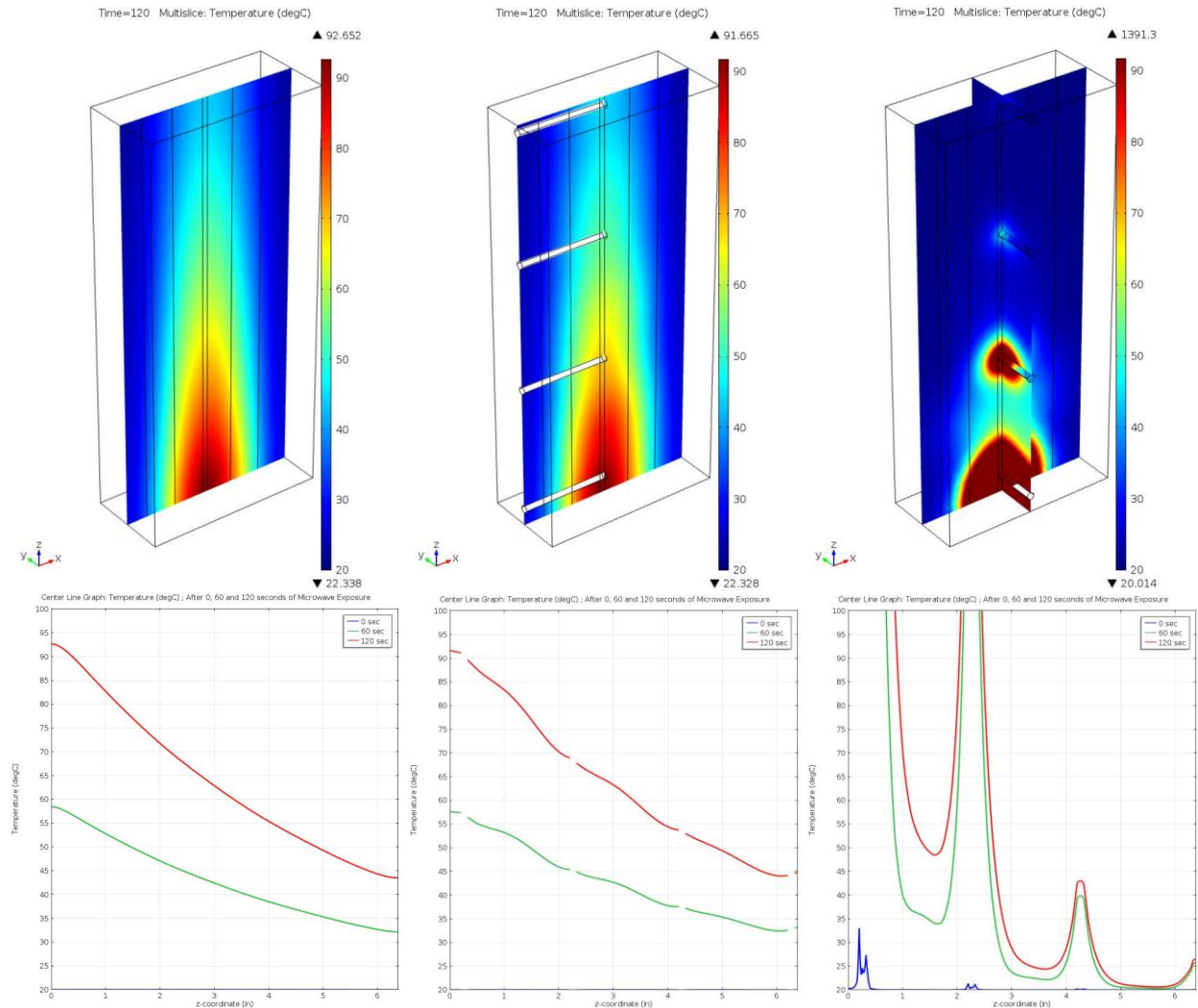
Figure 4. Feasibility of inserting temperature probes to waveguide center. Top Images: (Left-) temperature profile with no probes; (Center-) temperature profile with probes through center of narrow-wall and (Right-) temperature profile with probes through center of broad-wall. Bottom Images: temperature measured along center-line of waveguide corresponding to images directly above each.

A multitude of models were prepared and evaluated within the first three months of this work to support the design effort. A sampling of a few others are shown in Figure 5. It is worth repeating that the successful completion of this 6 month hardware development project was, to a large degree, only made possible with the aid of computer modelling and rapid evaluation of multiple design options.
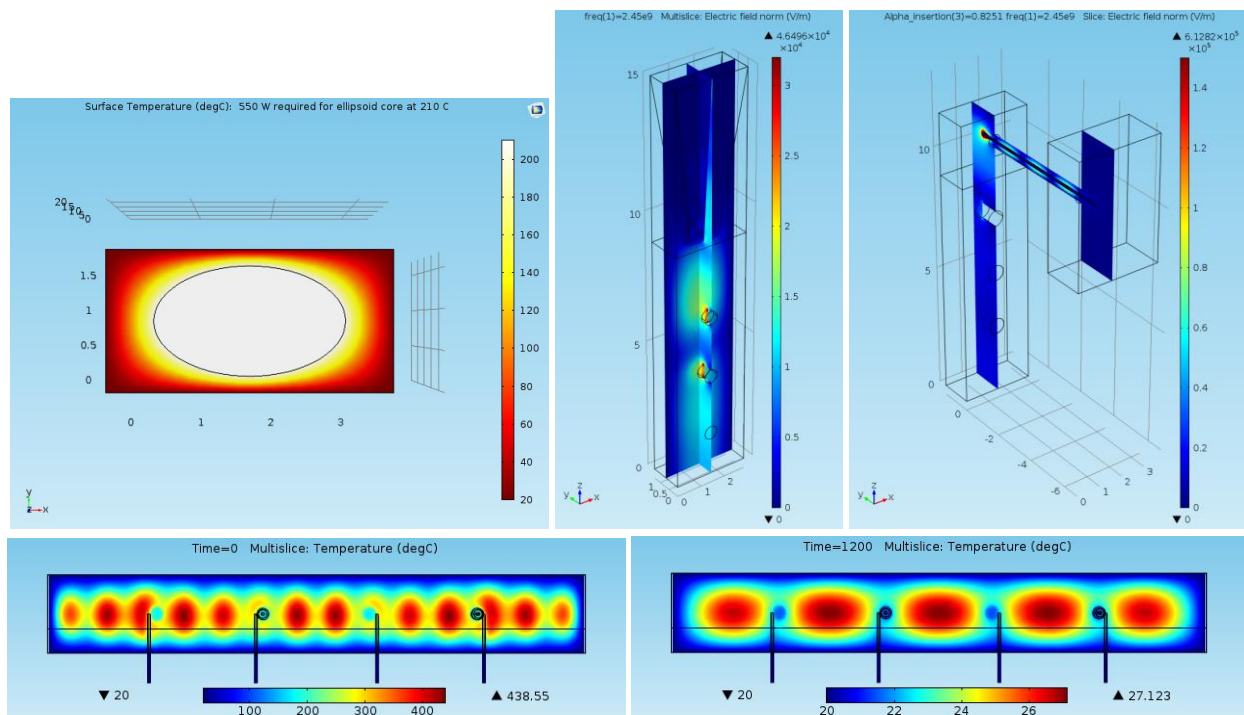
Figure 5. Various MRSHP development models. <u>Top Images</u>: (Left-) Estimation of power requirements required heating required media mass; (Center-) design verification of a custom 3-stub tuner and (Right-) full modeling of tuning and power transfer through a coaxial cable to a waveguide based load. <u>Bottom Images</u>: Temperature profiles (ºC) of (Left-) sorbent bed after 20 minutes microwave heating at 400 W; and (Right-) sorbent bed after 20 minutes subsequent ambient cooling when microwave power removed.

V.   Sub-Scale Prototype Design and Testing

Initial microwave regenerable bed design work looked at the fundamental feasibility of using computer modeling to introducing microwave power to a hollow WR340 waveguide via a coaxially fed antenna. While viability of this power transfer approach (coax to waveguide transition) is a well-established in real world applications, it was unclear whether the Comsol multiphysics modeling software available to us would serve in this design work. For initial evaluation a 0.125 inch diameter antenna was modeled as the simple extension of the inner conductor of a coaxial panel mount connector with PTFE as its dielectric. In this parameter optimization study, both the length and diameter of the antenna were varied over a range of values to identify its optimal size for maximum power transfer into the waveguide. In addition, by varying the position of the antenna relative to a back wall metal reflection plate, a maximum power transfer of 99.4% was attained. With the addition of 13x media to the waveguide volume an optimal power transfer of 97% was demonstrated. This approach, of constructing progressively more elaborate integrated models from stand-alone models, has been employed successfully thus far in our design work. Following this logical progression allows our models to gradually incorporate important aspects of an evolving design. While in and of themselves these particular models do not embody all the aspects of the final design, what is of primary importance, is the fact that their progressive successes demonstrate the viability of a computer modeling approach using the Comsol platform.

Models of both WR340 and 3 inch circular waveguide based sorbent bed designs were prepared and evaluated as depicted in Figure 6. For comparison sake, these beds were of the same internal volume. Each contained four coaxial antenna power feeds and four thermocouples with identical configurations. Other similarities include

equally spaced 0.125 inch diameter antennae that are each sheathed with their own 0.5 inch o.d. quartz tube. The electric field intensity pattern that develops within these waveguides at is shown at 100 W of 2.45 GHz power. Also shown is introduced on each cable. Antenna lengths were optimized to deliver maximum power to the sorbent loads (97% attained for the WR340 design and 94.5% for the 3 in circular design).
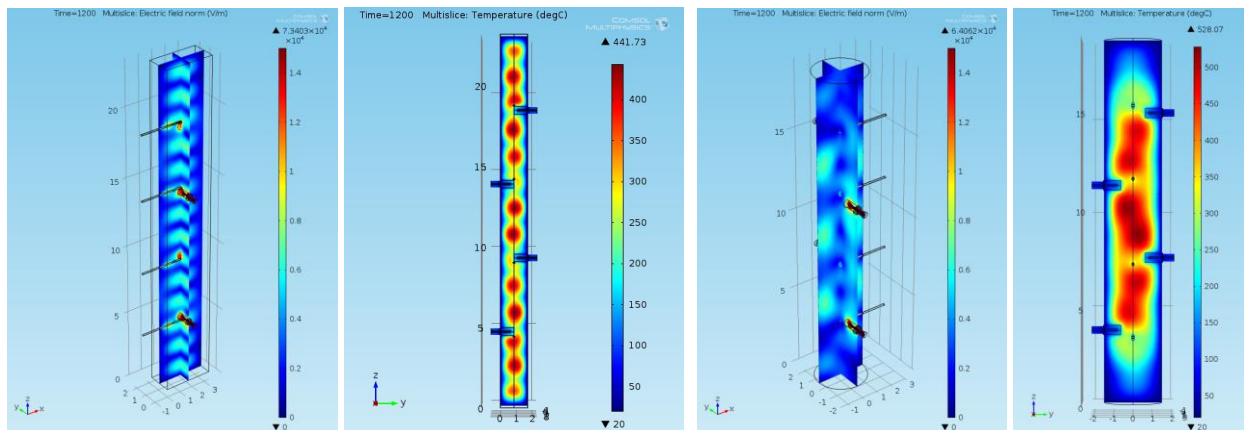
Figure 6. Computer Model Preparation using Comsol Multiphysics Software. (Two images on Left-) for WR 340 rectangular waveguide design and (Two images on Right-) for 3 inch circular waveguide design. Plots are of predicted electric field distribution and resulting temperature profile in media, respectively.

Figure 7 contains the 210 °C isothermal temperature profiles for both the WR340 and 3.0 inch circular waveguides that develop after heating nominally at 400 W for 20 minutes. The outer edge of these contours is at 210 °C, with temperatures above this at internal regions. Both waveguide designs do a good job of bringing the core temperature of the sorbent bed above minimum effective regeneration temperature. From a flow dynamics standpoint this is good since the majority of gas flow will pass through the cross-sectional center of the bed with reduced flow nearer the walls. For the circular waveguide, 41% of media volume is above 210 °C following 20 minutes of heating (at 385 W = 400 W @96% power absorption) versus only 29% of media exceeding 210 °C for the equal volume WR340 sorbent bed (at 378 W = 400 W @94.5% power absorption). These calculated percentages are strictly related to the volume of the media at or above 210 °C relative to the total bed volume, as depicted by either the box or cylindrical geometric outlines seen in Figure 7. Clearly, if all other considerations are equal, then the circular design is superior to the rectangular design in that a higher percentage of media volume participates in regeneration, thus giving more effective sorption capacity per unit bed volume.

While the circular waveguide may offer the potential for improved regeneration performance its geometry poses an inherent limitation on its cool-down rate due to the longer average thermal conduction pathway. As such, the duration of the cool-down phase following a sorbent bed regeneration is predicted by the model to extend longer than the targeted 30 minute rapid cool down interval. In addition, the fabrication length of a circular waveguide internal dimension within a box shaped exterior is limited by tool reach using EDM techniques. The length limitation for this type of waveguide is approximately 12 inches utilizing the same outside machine shop used during PPA fabrication. This is significantly less than the 19 inch circular waveguide bed that is required to match the bed volume of the WR340 waveguide bed design. Furthermore, cooling of a cylindrical bed is negatively impacted by the added thermal mass contained in the body material (aluminum) of the required box exterior. While in-wall cooling would significantly improve the cool-down rate, the aluminum volume added by the thick walls (particularly in the corners) would also greatly increase the overall bed mass. Importantly, the implementation of in-wall cooling channels greatly adds to the bed's complexity and cost of fabrication. Whereas external cooling plates keep complexity down and provide a measure of flexibility that may be needed if, for example, during testing the design is determined to need fine adjustments to achieve targeted performance. In an advanced scale-up design effort, with a lengthier development schedule and commensurate funding, utilization of a circular cross-section waveguide may be reasonably pursued. For the current sub-scale MRSHP prototype design however, a WR340 rectangular waveguide based sorbent bed was employed.
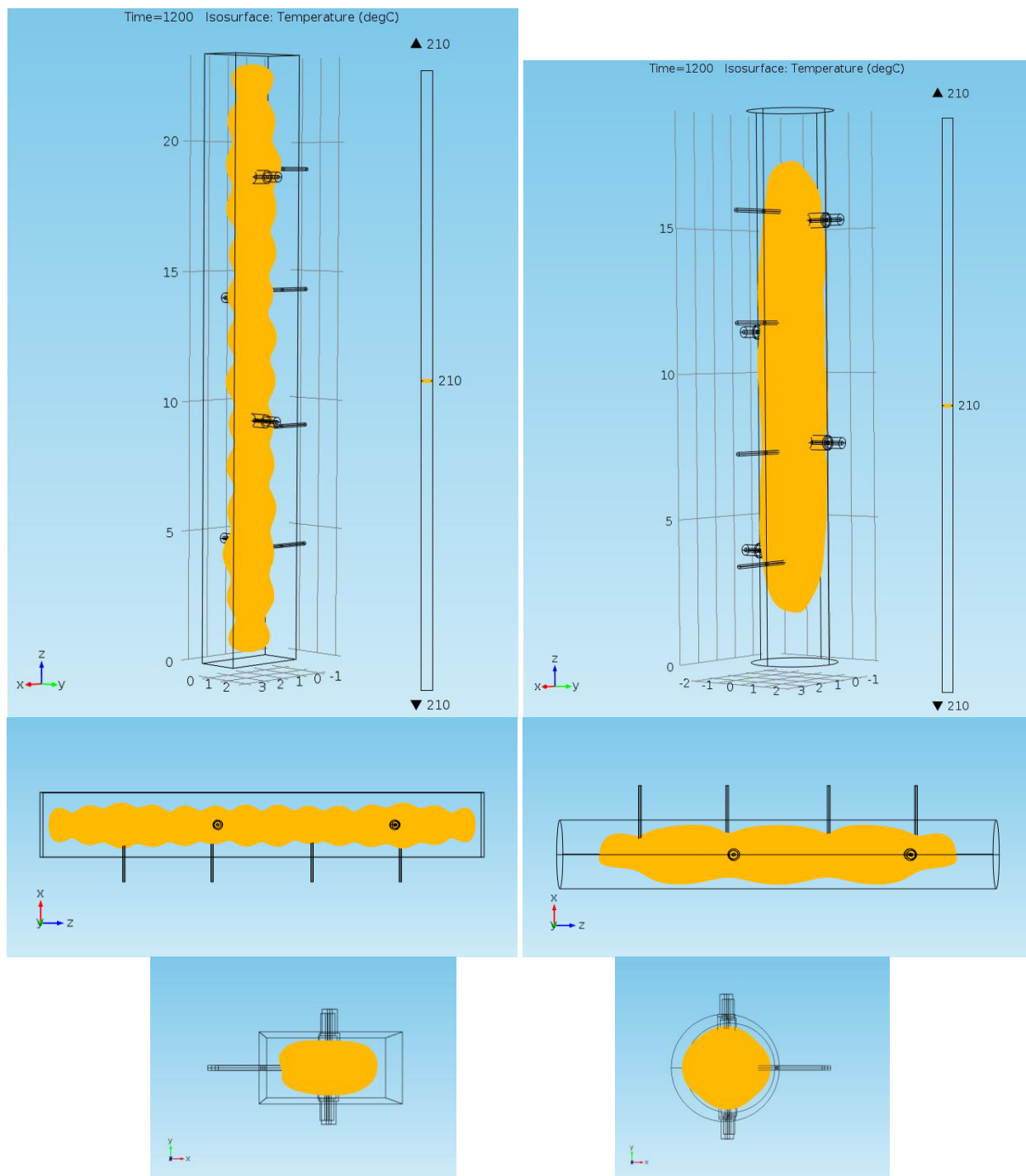
Figure 7. Isothermal 210 ºC surface plots after 20 minutes of 400 W heating using coaxially fed (4 ports) comparing WR340 rectangular (Left) and 3.0 inch i.d. circular (Right) waveguide bed designs. In each case thermocouples are present and walls are held at 20 ºC. Left Images are side profile (upper), broad wall (middle) and end (lower) views of the rectangular bed. Right Images are side profile (upper), broad wall equivalent (middle) and end (lower) views of the circular bed.

A process/hardware drawing of the Microwave Regenerative Sorbent Hydrogen Purifier (MRSHP) Sub-Scale Prototype Design is shown in Figure 8. This drawing shows some of the hardware interconnectivity detail along with pertinent process flow interfaces. Important attributes of the design include:

- Minimalistic hardware design for simplicity, compactness and reduced cost.
- External vacuum source, cooling water loop and gas flow control required.
- Distributed microwave power to media using coaxial cables.
- 1.2 kW continuously variable microwave power source with remote control capability using a 0-10 vdc "microwave adjust" input signal and an on/off 8-30 vdc "microwave enable" control voltage.
- Construction of microwave power elements using WR284 waveguide.
- Utilization of an E-Plane 90º waveguide elbow to locate half of waveguide elements in the lower portion of the rack and promote device compactness.
- Balanced microwave power transfer, using a H-pattern distribution structure formed from 3 waveguide T elements and 4 independent 3-stub tuners, with 4 waveguide to coax transitions.
- 4 low loss coaxial cables transfer 2.45 GHz regeneration power to the 13x bed.
- Display of input and reflected microwave power levels on dedicated meters (each with 0-1 v analog output).
- Thermocouple reader display of bed temperatures (has RS232 output).
- Loose containment of the sorbent bed and all gas connections within a LEL monitored hazardous gas enclosure.
- LEL alarm to remove gas flow and microwave power.
- External cooling plates located on the rectangular outer surfaces of a 23.3 inch long WR340 waveguide based media bed.
- Complete solenoid valve control of all inlet and outlet flows, for both gas and cooling water.
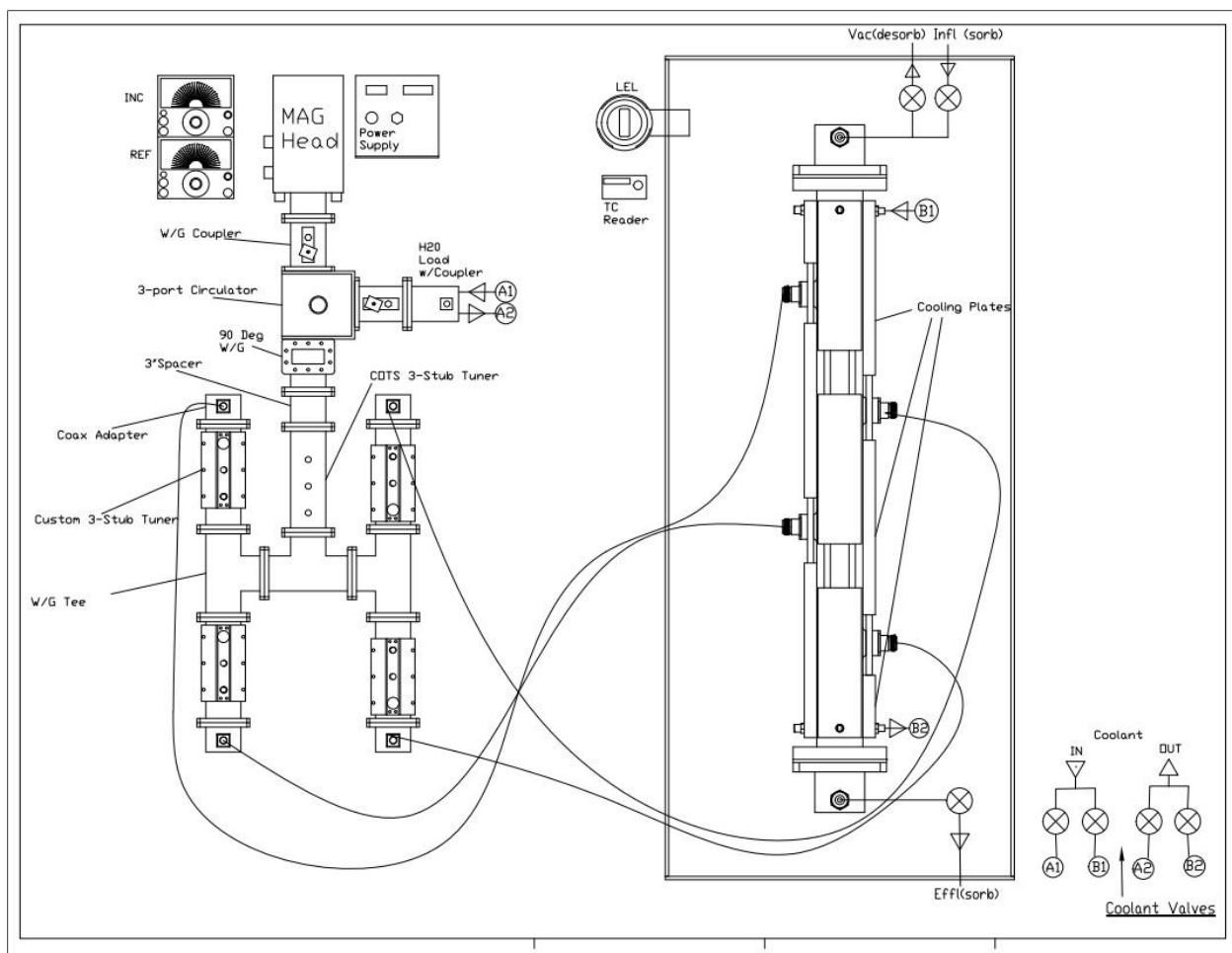
Figure 8. Microwave Regenerative Sorbent Hydrogen Purifier (MRSHP) Sub-Scale Prototype Process/Hardware Design. The Hazardous Gas Enclosure (HGE) is shown on the right with LEL monitoring.

Testing of the prototype was to entail performing multiple back to back sorption/desorption cycles demonstrating restored sorption capacity from one cycle to the next. However, due to damage resulting from operation at what proved to be excessive regeneration power and temperature levels (400 W, 300 °C), the bed-end coaxial cables connectors and sorbent bed antennae experienced some form of thermal runaway damage and required last minute replacement. Consequentially, due to time constraints, only a very conservative single microwave regeneration was performed at the greatly reduced conditions of 200 W, 210 °C for a 2-hour vacuum regeneration without damage to the device. This regeneration demonstrating a restored sorbent capacity that could operate for 8 hour at a 1-CM processing rate. Clearly, the regeneration time can be reduced by operation at higher power levels, yet remaining well below those that damaged the device. While not explicitly evaluated in this work, it is expected that a 1-hour vacuum regeneration can be safely achieved at some intermediate power and temperature levels (say 300 W, 240 °C, for example). In addition, as discussed earlier, modeling predicts that even if we could safely operate at 400 W, only 29% of the media reaches effective regeneration temperatures $\geq$ 210 °C in the WR340 waveguide. As such, much room for improvement remains to bring more of the media volume to into play. Again, this can in theory be achieved by instead using a circular cross-section waveguide bed which clearly merits thorough investigation in future work.

Several photos of the prototype are included in the figures in the Appendix. These include internal, side and back views of the device (Figure A1), as well as photos of the sorbent bed during media loading (Figure A2) and images of the system during final assembly and testing (Figure A3).

VI. Future Work

While showing great promise, a significant evolution must occur to bring this technology to a practical scale in order to support 4-6 CM operation of a full-scale PPA. Clearly, even though the successful outcome of this Phase 3 effort is very encouraging, further investment in the MRSHP development is required to achieve flight-ready hardware status in a timely manner to prepare for NASA's manned mission to Mars objective in the 2035 time-frame. The MRSHP prototype was designed to be a development unit that would facilitate further advancement of the technology. This is to be partially realized through independent testing at MSFC via integration with an actual PPA operating at a matched processing level (nominally between 1 and 2-CM scales). These test results would then logically feed into an advanced development effort performed at URC under either Phase 2 or advanced Phase 3 funding mechanisms.

VII. Conclusion

MRSHP technology has been significantly advanced by the development of a subscale prototype during an intensive 6-month Phase 3 effort. This work employed the results of a Phase 1 SBIR demonstration of feasibility effort and an aggressive development schedule to achieve this normally unrealistic objective. A mixture of computer modeling and select empirical testing was used in conjunction with parallel process design and hardware acquisition efforts to bring this project to a successful, timely conclusion with creation of MRSHP prototype hardware. This prototype now resides at MSFC where, at the time of writing this paper, it is preparing to undergo integrated testing and comparison with competitive approaches for hydrogen purification of PPA effluent.

Appendix
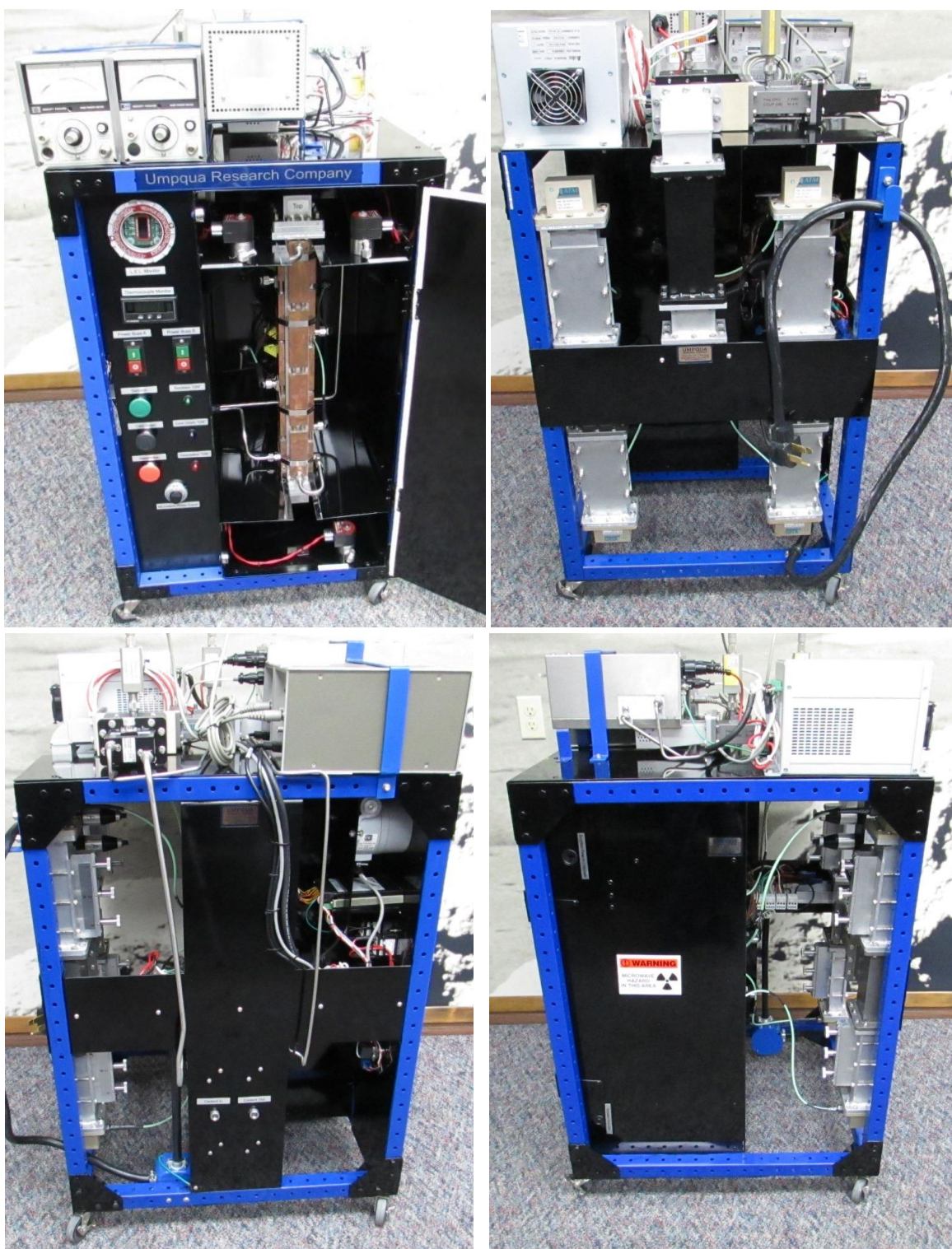
Figure A1. MRSHP Sub-Scale Prototype: (Upper Left-) Front view, door open; (Upper Right-) Back view; (Lower Left-) Left side view; and (Lower Right-) Right side view.

Figure A2. Loading of the WR340 rectangular waveguide based sorbent bed. Top to Bottom: (1st Row, Left-) Empty bed; (1st Row, Middle-) bed with quartz wool at bottom; (1st Row, Right-) First layer of 13x added; (2nd Row, Left-) Second layer added; (2nd Row, Middle-) Third layer added; (2nd Row, Right-) Final layer of 13x added; (3rd Row, Left-) Top layer of quartz wool added; (3rd Row, Middle-) Perforated plate added; (3rd Row, Right-) Gas inlet/distributer end piece added; (4th Row) Fully assembled bed. 2.2 L of Grace-Davison molecular sieve 13x media, received from Marshall Space Flight Center, was used to fill the waveguide based bed.

Figure A3. Preparation of the MRSHP prototype for delivery. Left to Right: (Upper Photos) Refurbishment-empty HGE, new cables w/ disassembled bed, media removed and saved; (Middle Photos) Reassembly- quartz sleeves affixed inside bed and shielding placed on thermocouple probes; (Lower Photos) Tuning, labeling, and preparation for shipping.

Acknowledgments

References

1. Holtsnider, J.T., Wheeler, R.R., Jr., Dewberry, R.H., Abney, M.B. and Greenwood, Z.W., Hydrogen Purification in Support of Plasma Pyrolysis of Sabatier Derived Methane, ICES-2014-195, presented 45th International Conference on Environmental Systems, Bellevue, July, 2015.
2. Wheeler, R.R., Jr., Hadley, N.M., Wambolt, S.R., Abney, M.B., Third Generation Advanced PPA Development, ICES-2014-0034, presented 44th International Conference on Environmental Systems, Tucson AZ, July 13-17, 2014.

3. Abney, M. B., Miller, L. A., and Barton, K., Evaluation of Sorbents for Acetylene Separation in Atmosphere Revitalization Loop Closure, AIAA Paper No. 2011-5057, presented at the 41st International Conference on Environmental Systems, Portland, July, 2011.

4. Atwater, J.E., Holtsnider, J.T., Wheeler, R.R., Jr., and Luna, B., Microwave-Powered Thermal Regeneration of Sorbents for CO2, Water Vapor and Trace Organic Contaminants, SAE 972430, presented at 27th International Conference on Environmental Systems, Lake Tahoe, NV, July 1997.

5. Holtsnider, J.T., Wheeler, R.R., Jr., and Dewberry, R.H., Compressorless Gas Storage and Regenerative Hydrogen Purification, Final Report Phase I, NASA Contract NNX14CM20P, Marshall Space Flight Center, December, 2014.

6. Abney, M. B., Miller, L. A., and Williams, T., Sabatier Reactor System Integration with Microwave Plasma Methane Pyrolysis Post-Processor for Closed-Loop Hydrogen Recovery, AIAA Paper No. 2010-6274, presented at the 40th International Conference on Environmental Systems, Barcelona, July 2010.

7. Mansell, J. M., Abney, M. B., and Miller, L. A., Influence of Oxygenated Compounds on Reaction Products in a Microwave Plasma Methane Pyrolysis Assembly for Post-Processing of Sabatier Methane, AIAA Paper No. 2011-5035, presented at the 41st International Conference on Environmental Systems, Portland, July, 2011.

8. Wheeler, R.R., Jr., Hadley, N.M., Dahl, R.W., Abney, M.B., Greenwood, Z., Miller, L.A., and Medlen, A., Advanced Plasma Pyrolysis Assembly (PPA) Reactor and Process Development, AIAA Paper No. 2012-3553, presented at the 42nd International Conference on Environmental Systems, San Diego, July, 2012.

9. Wheeler, R.R., Jr., Hadley, N.M., Dahl, R.W., Williams, T.W., Zavala, D.B., Jr., Akse, J.R. and Fisher, J.W., Microwave Enhanced Freeze Drying of Solid Waste, SAE 2007-01-3266, presented at 37th International Conference on Environmental Systems, Chicago, IL, July 2007.

10. Akse, J.R., Zavala, D.B., Jr., Wheeler, R.R., Jr., Dahl, R.W., Williams, T.W. and Griffin, D.W., Microwave Powered Gravitationally Independent Medical Grade Water Generation SAE 2007-01-3176, presented at 37th International Conference on Environmental Systems, Chicago, IL, July 2007.

11. Atwater, J.E., Akse, J.R., Wheeler, R.R., Jr., Dahl, R.W., Zavala, D.B., Jr., Hanken, B.E., Holtsnider, J.T., Williams, T.W. and Nanci, J.C., Hydrogen Recovery by ECR Plasma Pyrolysis of Methane, Final Report Phase I, NASA Contract NNM06AA45C, Marshall Space Flight Center, July, 2006.

12. Wheeler, R.R., Jr., Dahl, R.W., Hadley, N.M., Zavala, D.B., Jr., Williams, T.W., Akse, J.R. and Atwater, J.E., Microwave Enhanced Freeze Drying of Solid Waste, Final Report Phase II, NASA Contract NNA05CP02C, Ames Research Center, December, 2006.

13. Wheeler, R.R., Jr., Hadley, N.M., Dahl, R.W., Williams, T.W., Garmon, F.C., Akse, J.R., Atwater, J.E. and Fisher, J.W., Development and Testing of a Microwave Powered Solid Waste Stabilization and Water Recovery System, SAE 2006-01-2182, presented at 36th International Conference on Environmental Systems, Norfolk, VA, July 2006.

14. Akse, J.R., Zavala, D.B., Wheeler, R.R., Jr., PE, Dahl, R.W. and Williams, T.W., Microwave Powered Gravitationally Independent Medical Grade Water Generation, Final Report Phase I, NASA Contract NNC06CA77C, Glenn Research Center, 2006.

15. Wheeler, R.R., Jr., Dahl, R.W., Hadley, N.M., Williams, T.W., Garmon, F.C. and Atwater, J.E., Microwave Powered Solid Waste Stabilization and Water Recovery, Final Report Phase II, NASA Contract NNA04CA12C, Ames Research Center, 2006.

16. Wheeler, R.R., Jr., Atwater J.E. and Hadley, N.M., Microwave Enhanced Freeze Drying of Solid Waste, Final Report Phase I, NASA Contract NNA04CB24C, Ames Research Center, 2004.

17. Atwater, J.E., and Wheeler, R.R., Jr., Microwave Permittivity and Dielectric Relaxation of a High Surface Area Activated Carbon, Appl. Phys. A, 79, 125-129, 2004.

18. Atwater, J.E., and Wheeler, R.R., Jr., Temperature Dependent Complex Permittivities of Graphitized Carbon Blacks at Microwave Frequencies Between 0.2 and 26 GHz, J. Mater. Sci., 39 (1), 151-157, 2004.

19. Atwater, J.E., and Wheeler, R.R., Jr., Complex Permittivities and Dielectric Relaxation of Granular Activated Carbons at Microwave Frequencies between 0.2 and 26 GHz, Carbon, 41 (9), 1801-1807, 2003.

20. Atwater, J., Wheeler, R.R., Jr., Garmon, F., Microwave Powered Solid Waste Stabilization and Water Recovery, Final Report Phase I, NASA Contract NAS2-03124, Ames Research Center, 2003.

21. Atwater, J.E., Wheeler, R.R., Jr., Jovanovic, G.N., and Reed, B., Magnetically Controlled Microwave Powered Fluidized Bed Reactor, Final Report NSF Grant DMI-0128201, 2003.

22. Wheeler, R.R., Jr., Atwater, J.E., Akse, J.R., Holtsnider, J.T., and Luna, B., Development and Testing of a Microwave Powered Regenerable Air Purification Technology Demonstrator, SAE 2002-01-2403, presented at 32nd International Conference on Environmental Systems, San Antonio, TX, July 2002.

23. Atwater, J.E., Michalek, Wheeler, R.R., Jr., W.F., Dahl, R.W., Lunsford, T.D., Garmon, F.C., and Sauer, R.L., A Microwave-Powered Sterilizable Interface for Aseptic Access to Bioreactors That Are Vulnerable to Microbial Contamination, Biotechnol.Prog., 2001, 17, 847-851, 2001.

24. Sauer, R.L., Atwater, J.E., Dahl, R.W., Garmon, F.C., Lunsford, T.D., Michalek, W.F., Wheeler, R.R., Jr., Microwave Powered Sterile Access Port, U.S. Patent 6,077, 478, June 2000.

25. Atwater, J.E., and Wheeler, R.R., Jr., A Complex Permittivity Study of Sorbent Materials and Volatile Contaminants, Volume 1, Final Report, NASA Contract NAS2-97017, June 1999.

26. Wheeler, R.R., Jr., Thompson, J.O., Lunsford, T.D., Holtsnider, J.T., DeHart, J.L., Dahl, R., Akse, J.R., and Atwater, J.E., Development and Testing of the Microwave Regenerable Air Purification Technology Demonstrator, Volume 2, Final Report, NASA Contract NAS2-97017, June 1999.

27. Atwater, J.E., Dahl, R.W., Garmon, F.C., Lunsford, T.D., Michalek, W.F., Wheeler, R.R., Jr., and Sauer, R.L., Miniature Microwave Powered Steam Sterilization Chamber, Rev. Sci. Instrum., 68 (10), 3924-3925, 1997.