Particulate Filtration from Emissions of a Plasma Pyrolysis Assembly Reactor Using Regenerable Porous Metal Filters

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Microwave-based plasma pyrolysis technology is being studied as a means of supporting oxygen recovery in future spacecraft life support systems. The process involves the conversion of methane produced from a Sabatier reactor to acetylene and hydrogen, with a small amount of solid carbon particulates generated as a byproduct. The particles must be filtered before the acetylene is removed and the hydrogen-rich gas stream is recycled back to the Carbon Dioxide Reduction Assembly. We discuss developmental work on porous metal media filters for removing the carbon particulate emissions from the Plasma Pyrolysis Assembly exit gas stream and providing *in situ* media regeneration capability. Because of the high temperatures involved in oxidizing the deposited carbon during regeneration, there was particular focus in this development on the materials that could be used, the housing design, and heating methods. This paper describes the design and operation of the filter and characterizes their performance from integrated testing at the Environmental Chamber (E-Chamber) at MSFC.

Nomenclature

AES	=	Advanced Exploration Systems
CM	=	Crew Member
CRA	=	Carbon Dioxide Recovery Assembly
ECLSS	=	Environmental Control and Life-Support Subsystem
GC	=	Gas chromatograph
ISS	=	International Space Station
MSFC	=	Marshall Space Flight Center
PPA	=	Plasma Pyrolysis Assembly
SBIR	=	Small Business Innovative Research
SLPM	=	Standard Liters per Minute

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I. Introduction

PLASMA pyrolysis is being investigated as a candidate technology for recovery of hydrogen from the methane produced by the ISS Carbon Dioxide Recovery Assembly (CRA). In order to recover oxygen for continued ISS habitation, the CRA utilizes a Sabatier reactor that recovers oxygen from exhaled carbon dioxide in the form of water:

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$$

Future long duration missions will require reuse of as many resources as are possible to limit upmass and resupply. Current ISS systems dispose of the byproduct methane and resupply hydrogen for continued operation of the oxygen recovery system by the electrolysis of water. In order to recover some of the hydrogen, the Plasma Pyrolysis Assembly (PPA) partially pyrolyzes some of the methane from the Sabatier reactor waste stream with the dominant reaction being:

$$2CH_4 \rightarrow C_2H_2 + 3H_2$$

This hydrogen can be fed back into the Sabatier reactor and reduce the required water electrolysis necessary for system operation. Inefficiencies in the PPA process result in incomplete conversion of methane and side reactions, which produce additional hydrocarbons including ethylene and ethane. The PPA was developed under a NASA Small Business Innovative Research (SBIR) grant to assess this technology and is undergoing performance testing in NASA's Environmental Chamber (E-Chamber) at Marshall Space Flight Center (MSFC). [1-3]

UMPQUA Research Company, the SBIR company that developed the PPA, observed that 40 mg/hr of particulate carbon was produced during PPA operation.[1] This results in approximately 1 gram of carbon being introduced into the environmental system every day for a crew of four, leading to significant effects to downstream systems. Previous efforts have been made to characterize the emitted, fine dusty carbon particulate[4] and to evaluate HEPA, scrolling and a ceramic filter[5].

A porous sintered metal filter was developed, integrated with the PPA and tested at the MSFC. Design considerations and results of initial tests of the porous sintered metal filter are presented here.

II. Filter Design

In order to maximize filter lifetime while minimizing consumables, a porous sintered metal filter made from Hastelloy X was designed. This porous medium is formed from metal powders and has an interconnected pore structure, which is optimized and controlled using metallurgical and other fabrication practices. These filters, which offer high purity filtration and mechanical and thermal stability, are commonly used for in-line gas processing and in extreme environments. The porous media chosen for the present tests is made of 0.5 micron grade porous Hastelloy X media, i.e. a 0.5 micron average pore size. This pore rating can provide greater than 99% overall particle capturing efficiency for all particle sizes at a specified volumetric flux of 107 actual $m^3/hr/m^2$ (6 $acfm/ft^2$). Hastelloy X was chosen in particular to allow heating of the filter assembly to 788 ^oC, where material stability at these elevated temperatures and oxidative environments is required. Another key property is the favorable thermal conductivity offered by these metal alloys which could be used to facilitate thermal regeneration. The ability to conduct heat through and into the filter medium makes this an attractive approach for high temperature oxidative regeneration of the captured carbon. Therefore, metallic filters overcome the challenge of heating poorly conductive media such as ceramic monoliths.

The filter assembly is shown in Figure 1. The construction of the filter assembly consisted of five filter tube elements and a large circular mounting base. Each filter element is a 15.24 cm long, 0.635 cm diameter tube with 0.16 cm thick walls. The tubular design was chosen to maximize filter surface area among filter media shapes available from Mott Corporation, the media manufacturer. The cylindrical housing was chosen to maximize radiative and conductive heating during filter regeneration. The wall of each tubular filter element is made from the porous Hastelloy material, while the ends are capped at the tip with a welded solid piece of Hastelloy and open at the base end. The filter tubes have been welded onto the mounting base to ensure a good seal, while keeping the inside of the tubes open at this end. The through holes in the base are for supporting and mounting it to the housing and the perpendicular holes are for cartridge heaters that were not used for the described testing. The housing is shown in Figure 1 with the filter installed. A type K thermocouple was inserted through the length of the center filter element and placed in contact with the tip end.

The filter assembly was installed and sealed in a steel housing fashioned with inlet and outlet ports for the gas flow, and pressure taps to measure differential pressure. Initial tests were conducted in a 316 stainless steel housing,

and subsequently in a Hastelloy X housing when it became available (because of the long lead time in fabrication). The housing was wrapped with heating tape. The heater was wrapped in a blanket of quartz wool and fiberglass insulation placed around the whole filter housing to minimize heat loss and protect the operator from the high temperatures.

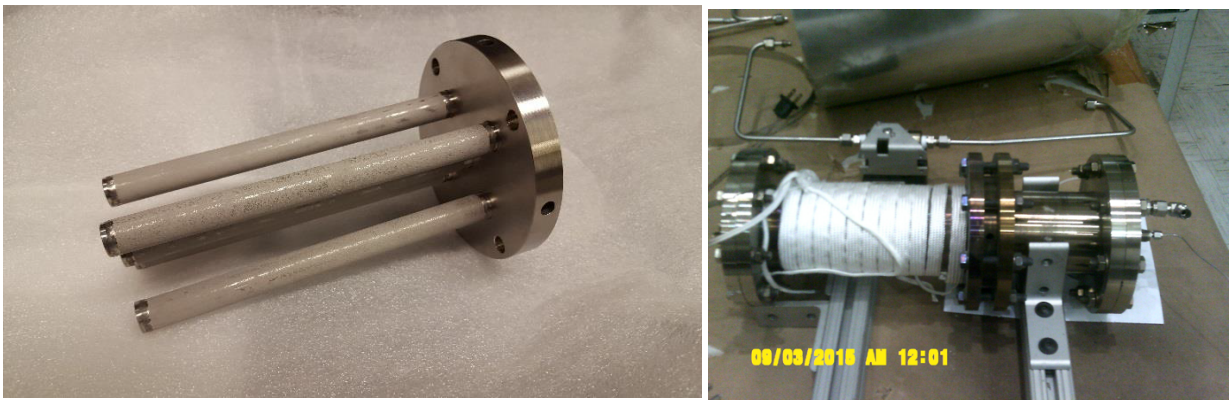

Figure 1. Left, the Hastelloy X filter element as received. Right, the filter mounted with heating tape for regeneration. The filter is inside the portion between the left and middle flange. Gas flow is directed onto the outside of the filter elements, from left to right in both images.

III. Test Setup

The PPA was integrated into MSFC's E-Chamber located in the MSFC ECLSS Development Facility. The E-chamber provides a testing architecture for integrating various life support systems hardware and provides the flight-like interfaces and process gas flows required to replicate the various life support processes on the International Space Station (ISS). The tests were conducted at a 4 crew-member (CM) flow rate equivalent to 8 Standard Liters per Minute (SLPM). The mixture of hydrogen, acetylene, methane and ethylene produced in the PPA was introduced into the filter housing where it first interacted with the outer surface of the filter elements and then flowed through the inner core channel.

The PPA gas flow was initiated under nominal conditions from either compressed gas cylinders (1.4 SLPM methane and 5.6 SLPM hydrogen at 110 torr) or directly from an operating CRA ground development unit. The product gas mixture containing carbon particulate was fed into the porous metal filter until the pressure drop from the filter doubled. A HEPA filter, described in previous studies,[5] was installed downstream of the experimental filter as a measure to protect downstream components from particulate not captured in the experimental filter.

Regeneration of the filter element can theoretically be accomplished by elevating the filter temperature and diverting CO_2 from the CRA inlet through the PPA system and into the porous metal filter to clean solid carbon from the filter surfaces using the reverse Boudouard reaction:

$$C + CO_2 \rightarrow 2CO$$

The byproduct CO would then be removed from circulation as a waste product. No plans exist to recover this minimal amount of oxygen, a sacrifice to continued ECLSS operation.

Heating for regeneration was applied through the heater tape on the outside of the housing using a microprocessor based controller to raise and maintain the internal filter temperature to 500-600^{o}C. The heating tape is turned off during normal operation of the PPA. The gas conditions remained at 110 torr during regeneration but gas flow was set to 8 SLPM of CO_2. A gas chromatograph (GC) calibrated with gas standards for the gases associated with PPA function was used to measure the gas concentrations of CO and CO_2 to monitor the progress of oxidative filter cleaning. The heater tape was heated to achieve 550 ^{o}C at the closed tip of the middle filter element. The PPA continued to be powered to preheat the CO_2 gas stream flowing into the filter housing. Regeneration was carried out for 4-6 hours to ensure that the filter was cleaned, even though this was beyond the point when the GC measurement of CO had decreased to a steady concentration.

IV. Initial Results

After about 20 hours of nominal operation at the 4-CM flow rate, the filter surfaces became loaded with a layer of carbon particles emitted by the PPA reactor. The particulate carbon build up on the exterior of the porous metal filter is shown in Figure 2, which shows an early borescope image of the interior of the filter housing. A uniform layer of carbon deposit is clearly seen on all the exposed tube surfaces with minor clear patches where the bare porous metal is seen. The carbon deposits even extend to the base of the filter assembly and on the inside walls of the housing (not shown in the figure). Therefore, all internal surfaces in contact with the carbon particle laden flow are potentially carbon capturing surfaces. Consequently, during regeneration the housing as well as the filter assembly will require heating to remove the built-up carbon. A small trace amount of carbon was seen through the sight glass on the downstream HEPA filter.

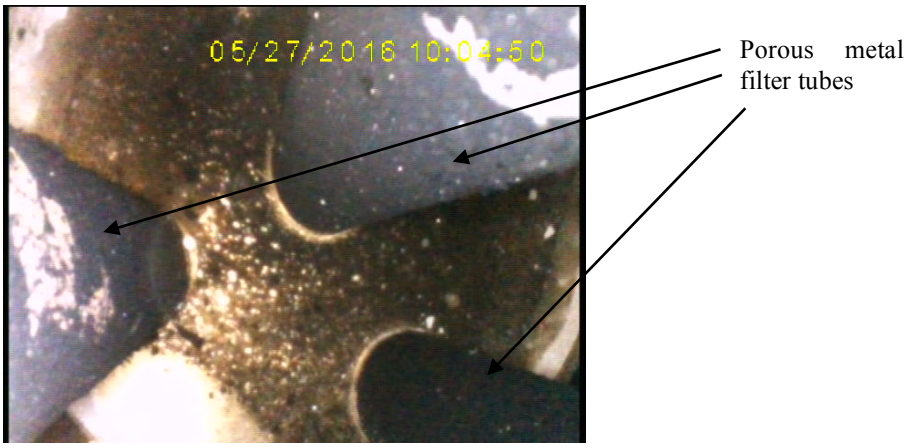

Figure 2. A borescope image of carbon particulate collecting on the porous metal filter elements within the filter housing – 3 of the 5 filter elements are visible. The carbon deposit can be seen covering most of the filter tubes and the base of the assembly.

The pressure drop data across the test filter was initially measured using a 1 PSID pressure transducer which was subsequently found to be faulty. As a result, system pressure data was alternately used as a measure of the pressure drop across the filter. The difference in pressures between the PPA reactor and the back pressure measured at the vacuum pump inlet provided the pressure drop across the test filter and secondary filter elements installed downstream of the test filter. A test run was also conducted without the test filter to provide the pressure drop contribution from the additional filter components and the system plumbing, which was used to calculate the pressure drop solely across the test filter.

Figure 3 provides a representative plot of absolute system pressures and filter pressure drop during one of the runs. The plots of pressure show some variability with time. The extended testing over tens of hours shown in the plot required that testing be performed over several days. Some of the variability in the plots are attributed to this operational sequence, particularly the drop-offs and sudden dips in the data. In general, particulate loading on the filter was characterized by a constant rise in pressure drop. The rate of pressure drop rise, which is generally linear, can be used to gauge the operational cycle of the filter between regenerations. Typically, a media filter is allowed to reach twice its initial pressure drop rise before it needs to be replaced or reconditioned. Because of the mechanical strength and stability of the porous metal filter an excess of above twice the initial pressure drop may be considered if it still meets system requirements. Subsequent tests after regeneration showed that in general the pressure drop is able to recover back to its base value.

This level of pressure drop shown in Figure 3 is typical for fine pore porous media in gas flows. Although the PPA is able to operate at this level of pressure drop, it may prove excessive for a flight system. Therefore, a critical area of

improvement will be to reduce the pressure drop through design changes. This may include increasing the total filter surface area by additional tubes, larger diameter tubes and larger media pore rating.

The gaseous effluent constituents were monitored with the GC during regeneration. As the filter is heated to its set point of 500 to 600 °C, the levels of CO_2 (oxidizing gas) and CO (product gas) indicated the level of regeneration taking place. Figure 4 shows the plot of the GC data for a typical 6 hour regeneration event. The gas concentrations appears to peak after the start of regeneration and then level off after approximately an hour, as the temperature and oxidation reaction reach equilibrium.

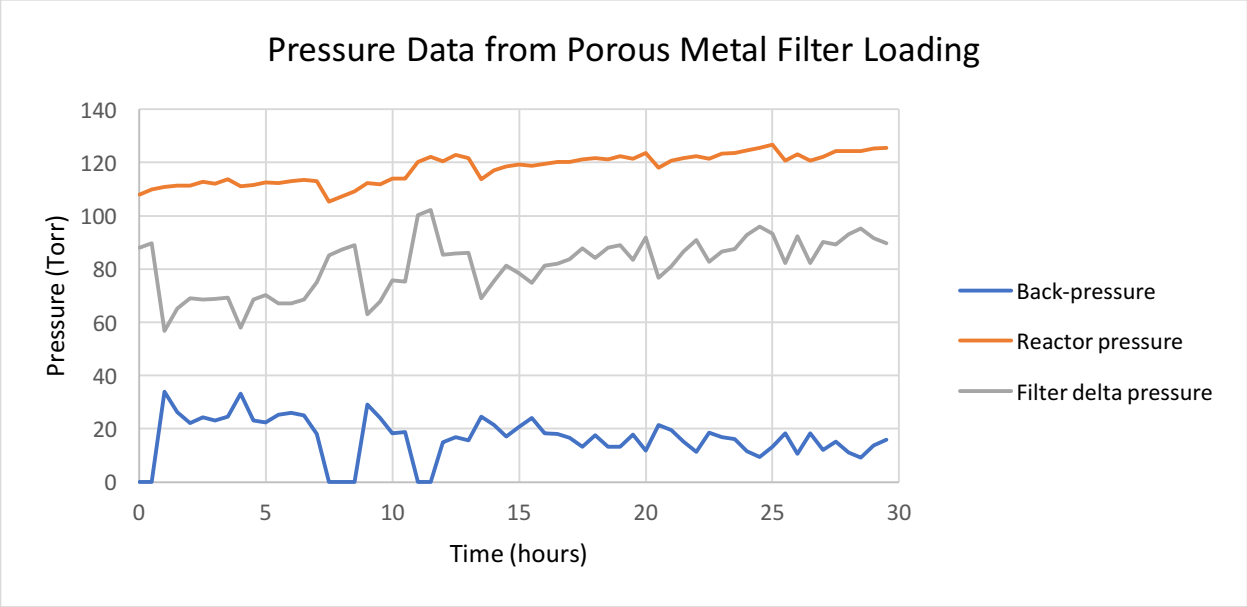

Figure 3: System pressures and filter pressure drop measured during nominal PPA run.

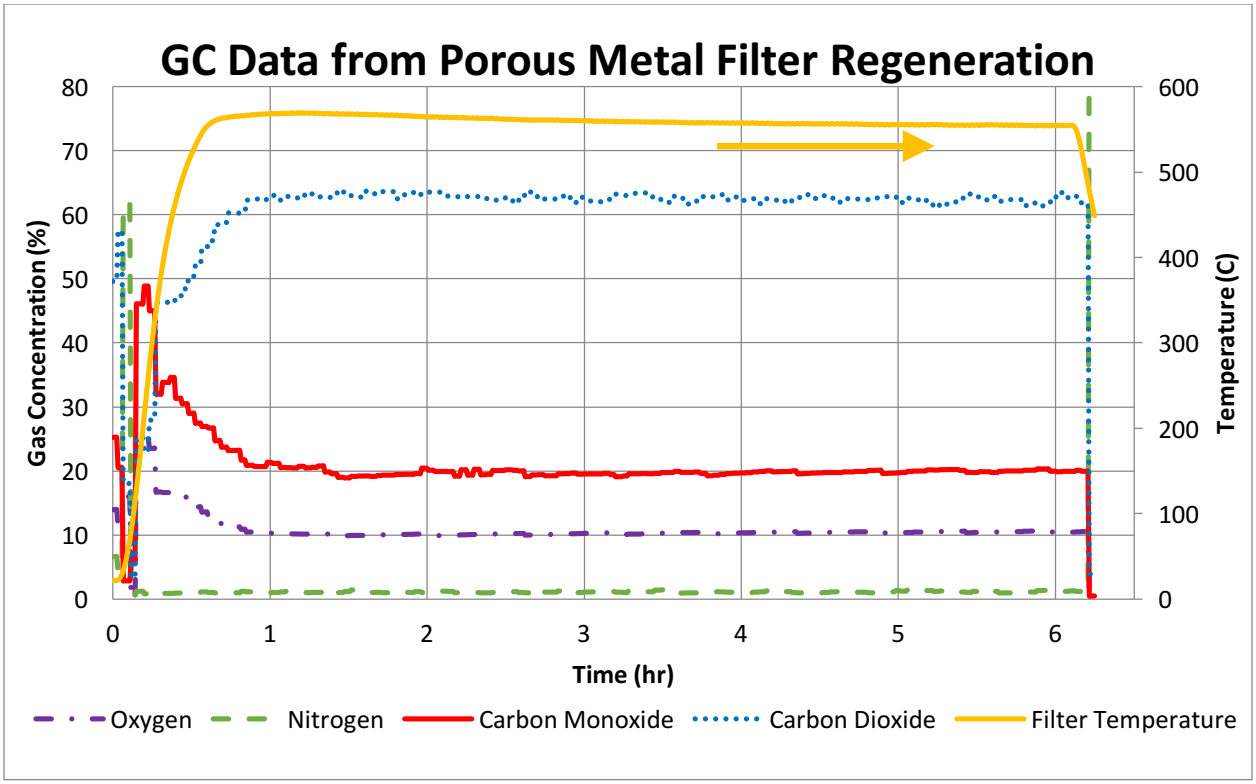

Figure 4: Gas chromatograph data from regeneration of porous metal filter. Carbon dioxide was passed through the PPA at a reduced power before introduction to the heated porous metal filter to remove elemental carbon as carbon monoxide. This example run shows the highest concentrations of carbon monoxide and oxygen at the initial injection of carbon dioxide before the filter came to regeneration temperature (550^{o}C). The original intent was to use the PPA to preheat the incoming gas but the plasma also appears to dissociate some of the carbon dioxide into oxygen and carbon monoxide.

V. Conclusions and Future Work

A porous sintered metal filter made from Hastelloy X was developed and tested in the MSFC E-Chamber. Initial tests show that the porous metal filter successfully collected particulate carbon from the PPA gas stream. Although most of the carbon is collected on the filter, a small trace amount of carbon was seen through the sight glass on the downstream HEPA filter. The particulate loading was characterized by a constant rise in pressure drop. CO_2 oxidation of solid carbon between 500-600 ^{o}C successfully returned the pressure drop over the filter to the nominal beginning level. This was corroborated by GC measurement showing the decline of CO produced through oxidation of carbon particulate.

Future efforts will focus on optimizing the filter. The PPA requires regeneration approximately every 6-8 hours to remove carbon deposits from the reactor stubs, though this typically takes only 15 minutes. Filter size and shape will be optimized for easier heating and heating elements that are able to provide higher temperatures will be integrated in order to determine whether this brings regeneration times into line with PPA regeneration. The size of the filter can also be decreased if necessary to aid this effort, though the filter may not need to be regenerated with each PPA regeneration. The pressure drop over the entire filter system also needs to be decreased by increasing the filter surface area, changing filter porosity or by other means to better match the current flight hardware design restrictions.

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