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PERFORMANCE ASSESSMENT OF BENCH-SCALE ELECTROTHERMAL SWING ADSORPTION PROTECTIVE FILTER SYSTEMS

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Electrothermal Swing Adsorption (ESA), or Joule-Effect heating of adsorbents in a cyclic adsorption process, is an energy-efficient technology on which to base a regenerative protective filter system. In an attempt to define the performance envelope of ESA systems, we evaluated three different ESA media: Activated carbon fiber cloth (ACFC), Carbon Fiber Composite Molecular Sieve (CFCMS), and Microlith™ (a trademark of PCI, Inc., a zeolite-coated wire mesh). Bench-scale systems based on these media were constructed and evaluated. Coincidentally, the three systems used different reactor morphologies: baghouse, axial flow, and radial flow. The performance of these systems in terms of removal efficiency, power consumption, weight, cube, and operability is discussed.

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Performance Assessment of Bench-Scale Electrothermal Swing Adsorption Protective Filter Systems

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ABSTRACT

Electrothermal Swing Adsorption (ESA), or Joule-Effect heating of adsorbents in a cyclic adsorption process, is an energy-efficient technology on which to base a regenerative protective filter system. In an attempt to define the performance envelope of ESA systems, we evaluated three different ESA media: Activated carbon fiber cloth (ACFC), Carbon Fiber Composite Molecular Sieve (CFCMS), and Microlith™ (a trademark of PCI, Inc., a zeolite-coated wire mesh). Bench-scale systems based on these media were constructed and evaluated. Coincidentally, the three systems used different reactor morphologies: baghouse, axial flow, and radial flow. The performance of these systems in terms of removal efficiency, power consumption, weight, cube, and operability is discussed.

INTRODUCTION

All fielded military Nuclear Biological Chemical (NBC) individual and collective protection systems utilize non-regenerative particulate and gas/vapor filters operated in single-pass mode. These collective protection filters can require frequent replacement of the filter elements. These filter change-outs are costly and add a significant logistic burden to field operations. Because there is no existing indicator of remaining carbon filter life, uncertainty can lead to premature filter change-outs. A regenerative filter system would be desirable as it would not only reduce the logistical burden of using consumable filter elements, but would also eliminate or greatly reduce the need of end-of-life indication.

Toxic Industrial Chemicals (TICs) can be used by terrorists, as a low-technology alternative to military chemical weapons. Many low-molecular-weight (LMW) TICs are weakly adsorbed by conventional activated carbons, and so in the current single-pass filters they are removed by chemisorption or by chemical reaction with impregnants added to the activated carbon adsorbents. One challenge in creating a regenerative filter system is to create sufficient capacity to capture TICs by physisorption or reversible chemisorption. Another challenge in creating a regenerative filter system is to thoroughly desorb strongly bound compounds such as organophosphates.

In an effort to assess the feasibility of current regenerative filter technologies, the Joint Service Chemical and Biological Defense Program (JSCBDP) funded a research project entitled:

"Advanced Regenerative Filtration Maturation for Collective Protection Applications". The advanced regenerative filtration project examined thermal, pressure, and electrical swing methods to regenerate adsorption-based filter systems for the removal of Chemical Warfare Agents (CWAs). The ESA (Sullivan, et.al. 2004) results using Activated Carbon Fiber Cloth (ACFC) were promising, so the JSCBDP funded a subsequent (this) project entitled "Evaluation and Maturation of ESA Regenerative Filter Media", wherein systems based on three different ESA media are compared. The other media that had been demonstrated in unrelated programs (Burchell, et.al. 1997), (Roychoudhury, et.al. 2004) to have potential for use in an ESA NBC filter included: Carbon Fiber Composite Molecular Sieve (CFCMS) and Microlith™ (a trademark of PCI, Inc., a zeolite-coated wire mesh).

Data such as breakthrough curves, equilibrium adsorption isotherms, and some desorption tests had been collected for all these media, but there is not sufficient information to design and optimize a full-scale prototype system. Testing at a systems level, with adsorption–desorption cycling, optimization of operating parameters and regeneration algorithms, and iteration on adsorber and adsorbent design was desired to advance ESA technology and to define the potential performance of ESA for collective protection. Each participant, AFRL, ORNL, and PCI, Inc. prepared two adsorbers to be operated in tandem in automated continuous operation. The participants attempted to design adsorbers optimized for pressure drop, weight, cube, and energy consumption for treating a 5 CFM flow. Herein we report some initial results for this effort.

EXPERIMENTAL

Equilibrium adsorption isotherm experiments were conducted with a Gravimetric Sorption Analyzer, Model GHP-F, manufactured by VTI Corporation of Hialeah, Fla. (Figure 1). The system uses a Magnetic Suspension Balance by Rubotherm of Bochum, Germany.

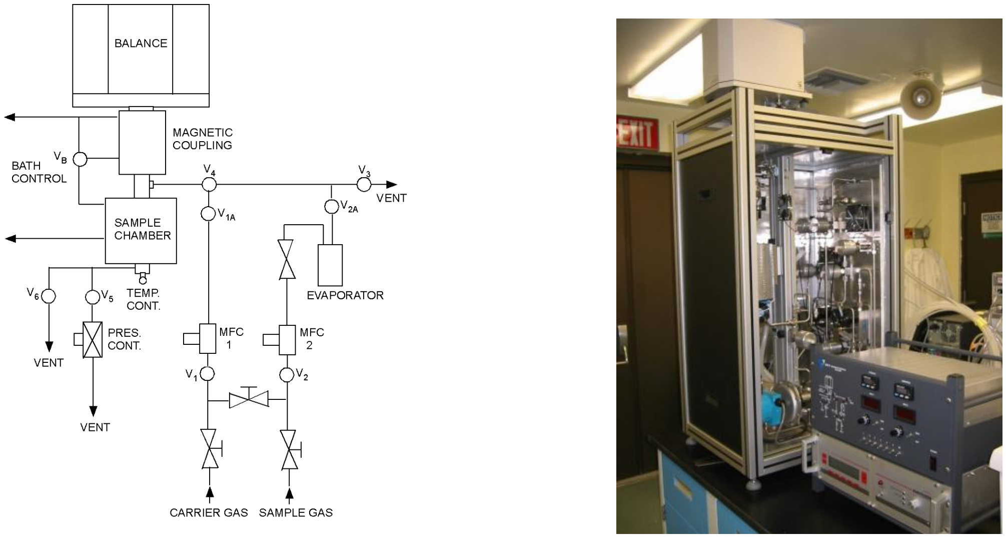

Figure 1. Gravimetric Sorption Analyzer Model GHP-F Schematic and Photo

The adsorbers were tested on a 5 CFM custom-built experimental test stand that consisted of gas driers, mass flow controllers, a water injector & vaporizer for humidification, control valves, a direct electrical heating power supply, gas detection units, and a distributed control system (National Instruments Fieldpoint™ hardware and Lookout™ software). Gas detectors used were either a Baseline-Mocon, Inc. Series 8800 Total Hydrocarbon Analyzer, or an Innova Photoacoustic Analyzer, Model 1412.

Figure 2. Test Stand for Adsorption System Experiments with CFCMS Adsorbers Online

The systems can be operated as depicted in Figure 3, with two vessels working in tandem to provide continuous protection. Regeneration purge gas flow is in the opposite direction to the challenge gas. Treated air is used as the purge gas, with generally 20% to 35% of the flow being consumed by the regenerating filter. Nitrogen is also available for a “super purge” allowing higher regeneration temperatures for the carbon-based adsorbents.

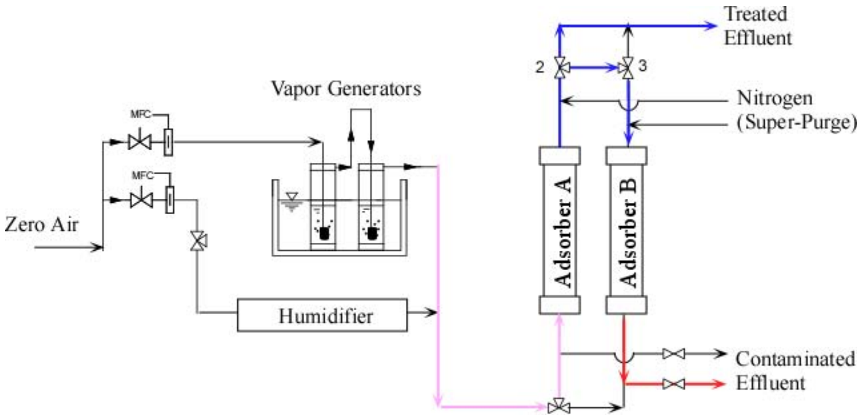

Figure 3: Test Stand Schematic, with the Adsorber "A" in Service and Adsorber "B" Regenerating

Breakthrough curves were collected for propane and R-134a. Adsorption capacity can be computed from the breakthrough curve by:

$$W = \frac{P_{tot}\,(MW)\,Q_{air}}{\rho_l m_s R T} \int_0^{t_{sat}} \left(\frac{C_{in}}{1 - C_{in}} - \frac{C_{out}}{1 - C_{out}} \right) dt \qquad (1)$$

where W is the volume of adsorbed chemical per unit mass of adsorbent, P_{tot} is the total pressure of the inlet gas stream, MW is the molecular weight of the adsorbate, Q_{air} is the volume flow rate of carrier gas, R is the ideal gas constant, T is the absolute temperature of the inlet carrier gas, m_s is the mass of the adsorbent, ρ_l is the bulk liquid density of the adsorbate, C_{in} is the mole fraction of adsorbate in the inlet gas stream, C_{out} is the mole fraction of adsorbate in the exhaust gas stream , and t_{sat} is the time at which C_{out} has reached its final, steady-state value because the adsorbent is saturated with respect to the adsorbate.

RESULTS

Capture of LMW TICs and addressing competitive water vapor adsorption were considered to be the most difficult challenges in achieving adequate performance. Equilibrium water adsorption isotherms were collected on the carbonaceous adsorbents in order to quantify water adsorption at various humidity levels to facilitate adsorbent selection and in order to explore strategies for operating in a high humidity environment. Chemical treatments of ACFC, including reduction with hydrogen (to increase hydrophobicity) and oxidation (to enhance capture of base-forming TICs) were explored. The results of these experiments are documented more completely in *Adsorption* (Sullivan, et.al, 2007), but some examples are presented in Figure 4. Because the zeolites in the Microlith™ are relatively hydrophilic, PCI, Inc. chose to use a second water-removing adsorber upstream of the primary adsorber.

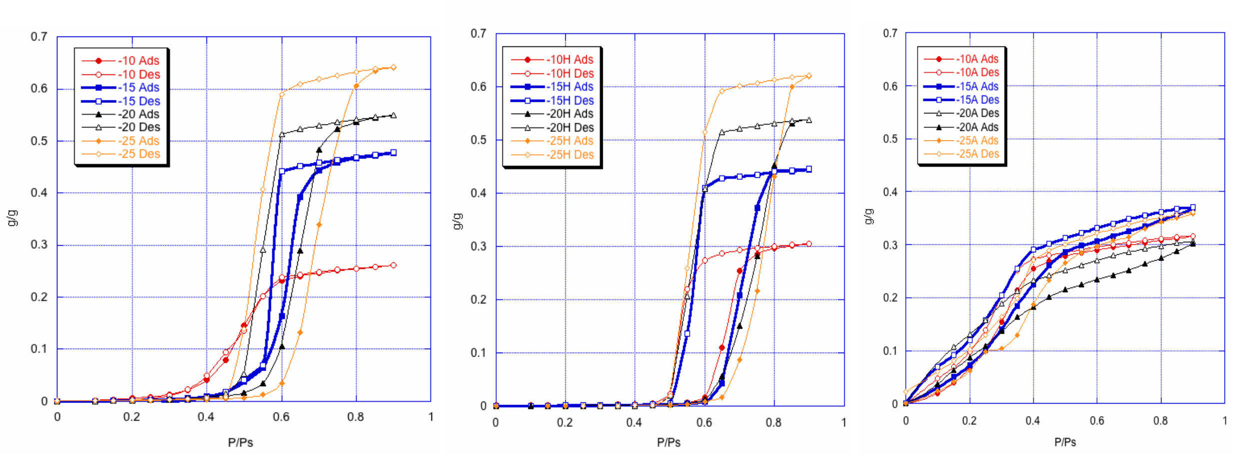

Figure 4: ACFC Water Adsorption: ACFC (left), H_2 Treated (Center), and Oxidized (Right)

More highly-activated ACFCs, and those treated with H_2, were found to be reasonably hydrophobic. With a modest temperature elevation of 5°C-10°C, which could be achieved by applying a modest current to the bed during adsorption, water uptake onto these ACFCs could be minimized. However, the highly-activated ACFCs are less effective at adsorbing LMW TICs. Treatments, such as oxidation, to improve TIC adsorption may have the unwanted side-effect of greatly increasing water adsorption.

In addition to the water vapor adsorption tests, equilibrium adsorption isotherms were collected for potential simulants for the TICs, including ethane, propane, Freon-23, and R-134a. HCN was considered to potentially be a design-limiting TIC, which is not well simulated by other compounds, so numerous equilibrium adsorption isotherms were also collected for this TIC. Most of the carbonaceous adsorbents performed poorly against HCN, with a general trend for higher adsorption with decreasing level of activation (and associated pore diameter). However, an electrospun activated carbon nanofiber (ACnF), manufactured by eSpin Technologies, Inc. of Chattanooga, TN, was found to have favorable, and most importantly, reversible HCN adsorption. The ACnF was not available in sufficient quantity to include in the ACFC adsorber, but this adsorbent could easily be layered with ACFC to create a composite bed.

The Microlith™ adsorbents initially had mediocre performance against HCN, on a weight basis, due to the low ratio of adsorbent to inert material of the metal framework. In response, PCI, Inc. developed a Microlith™ with a conductive glass core that greatly increased the ratio active material in the adsorbent.

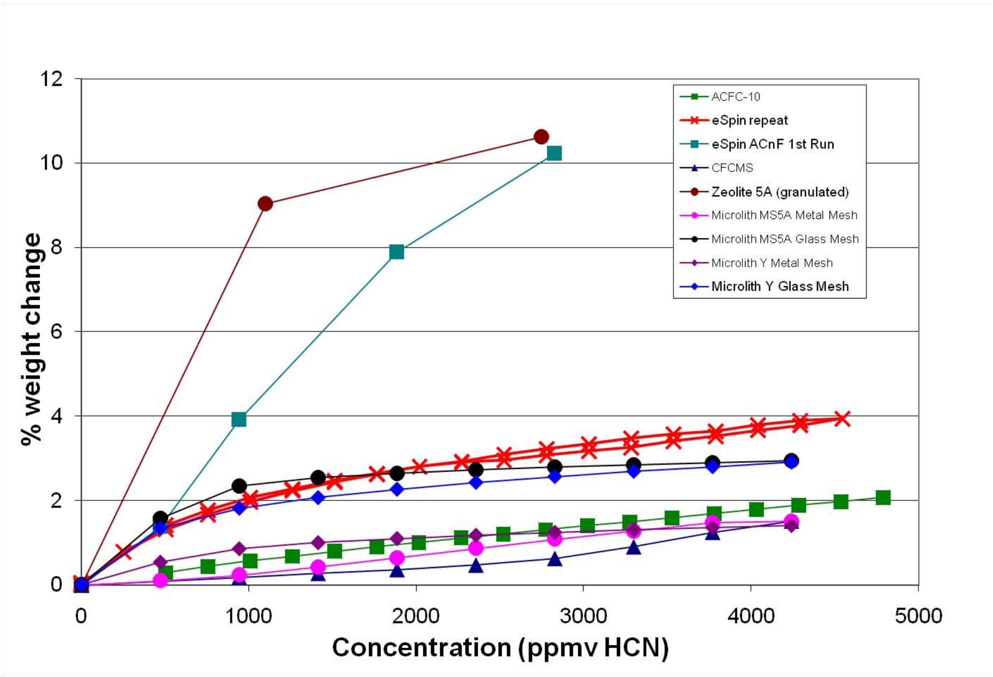

Figure 5: HCN Adsorption: The Highest Adsorption was Observed for Microliths™ and ACnF

Different reactor morphologies were selected by the designers as being best suited for the individual characteristics of each of the ESA media: a baghouse for the ACFC, an axial flow monolith for the CFCMS, and radial flow bed, or "jelly roll", for the Microlith™.

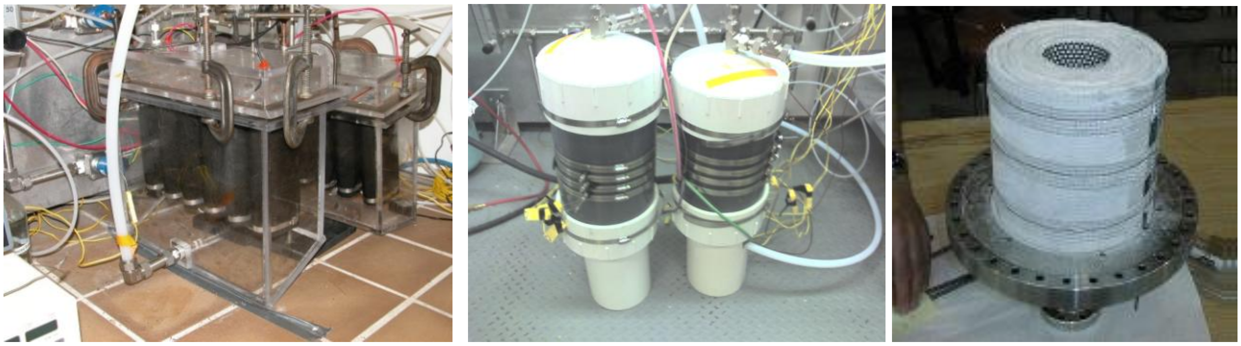

Figure 6: Adsorber Modules: ACFC (left), CFCMS (Center), and Microlith™ (Right)

Although one of the project objectives was to minimize weight and cube, each of the adsorber modules has significant potential for further optimization. The ACFC adsorber was constructed of 12mm acrylic so that it would be transparent and still withstand heating, but a thinner-walled aluminum construction would greatly decrease weight. The CFCMS adsorber has a significant volume above and below the monolith that could potentially be reduced. The Microlith™ modules would also benefit from an aluminum vessel. Also, it may be possible to integrate the water-removing adsorber into the primary unit.

Table 1: Dimensions of a Single Adsorber Module

		Adsorber (kg)	Adsorbent (kg)	Dimensions (m)	Volume (m^3)
Microlith™	Moisture	12.3	1.67	0.254 Dia × 0.216 Length	9.29E-03
	Contaminant	16.8	2.30	0.305 Dia × 0.216 Length	1.12E-02
CFCMS		9.5	0.72	0.241 Dia × 0.445 Length	2.03E-02
ACFC		11.8	0.60	0.406 × 0.387 × 0.152	2.40E-02

All of the adsorbers are of comparable volume, with a single unit being slightly larger than objective of 0.15 CF/CFM published in the FY10/FY11 DTRA Call for Proposals. Two adsorbers and the associated valving would thus be 2-3 times the DTRA goal at the 5 CFM scale. It is projected that a higher-flow system would have a lower specific volume than these prototypes, and that the DTRA goal would be achievable with further engineering.

Pressure drop of the Microlith™ adsorbers is much higher than that of the current fielded single-pass NBC filter set. Pressure drop of the ACFC system is acceptable. The CFCMS pressure drop should be reduced either by increasing the bed diameter-to-depth aspect ratio or making the monolith more porous.

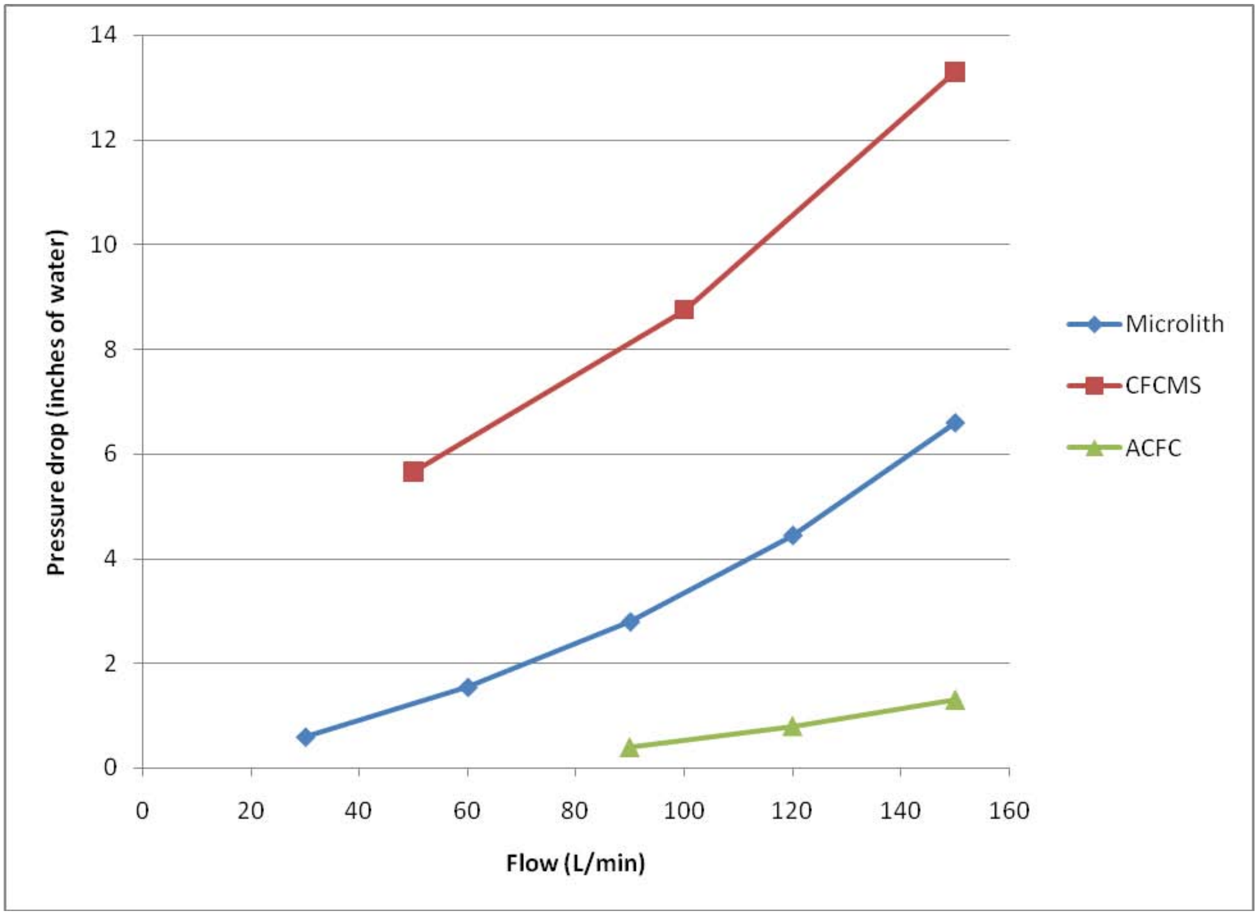

Figure 7: Adsorber Module Pressure Drop for ACFC, CFCMS, and Microlith™

Breakthrough curves for the systems against a propane challenge reveal that none of the systems are have ideal performance currently. The CFCMS appears to be experiencing significant channeling around the monolith and provides minimal protection in its current state. The Microlith™ has a high protection factor initially, but poor bed utilization. The ACFC has reasonable bed utilization, but is experiencing channeling or a leak, with about 1% of the challenge going unfiltered. Previous ACFC systems with similar configurations have not experienced this problem, so this appears to be a manufacturing defect rather than a design flaw.

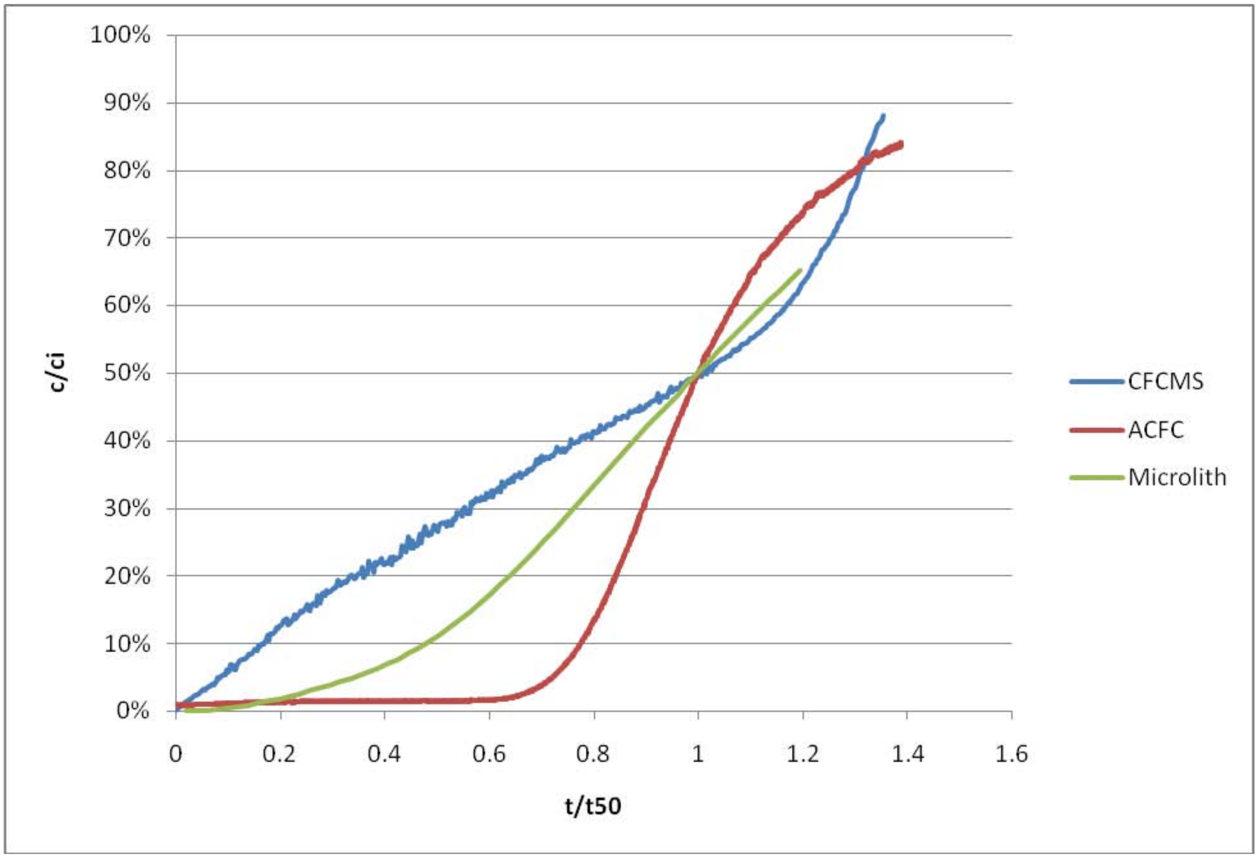

Figure 8: Propane Breakthrough Curves for ACFC, CFCMS, and Microlith™

The rate-limiting step of the cyclic adsorption process is the cooling of the adsorbent after regeneration, so examining the cooling cycle of a filter module is highly informative. The CFCMS filters experienced electrical shorting, so heating and cooling cycles could not be conducted on those units. A single heating and cooling cycle for the ACFC and the Microlith™ filters are compared (Figure 9). The Microlith™ filters have a higher regeneration target temperature (200°C), in order to fully desorb water and other contaminants. The ACFC can suffer oxidation at temperatures as low as 150°C, so a regeneration target temperature of 100°C was used. For the LMW stimulants, 100°C was adequate for complete desorption of the ACFC. The lower peak temperature of the ACFC allows more rapid cycling, which can result in either increased protection or reduced size of the unit. Additionally, the smaller adsorbent mass of the ACFC allows lower purge gas ratios.

A “super purge” at much higher temperatures could be used to more fully desorb heavier organics on an infrequent basis as a maintenance procedure, but would not be required in a threat scenario.

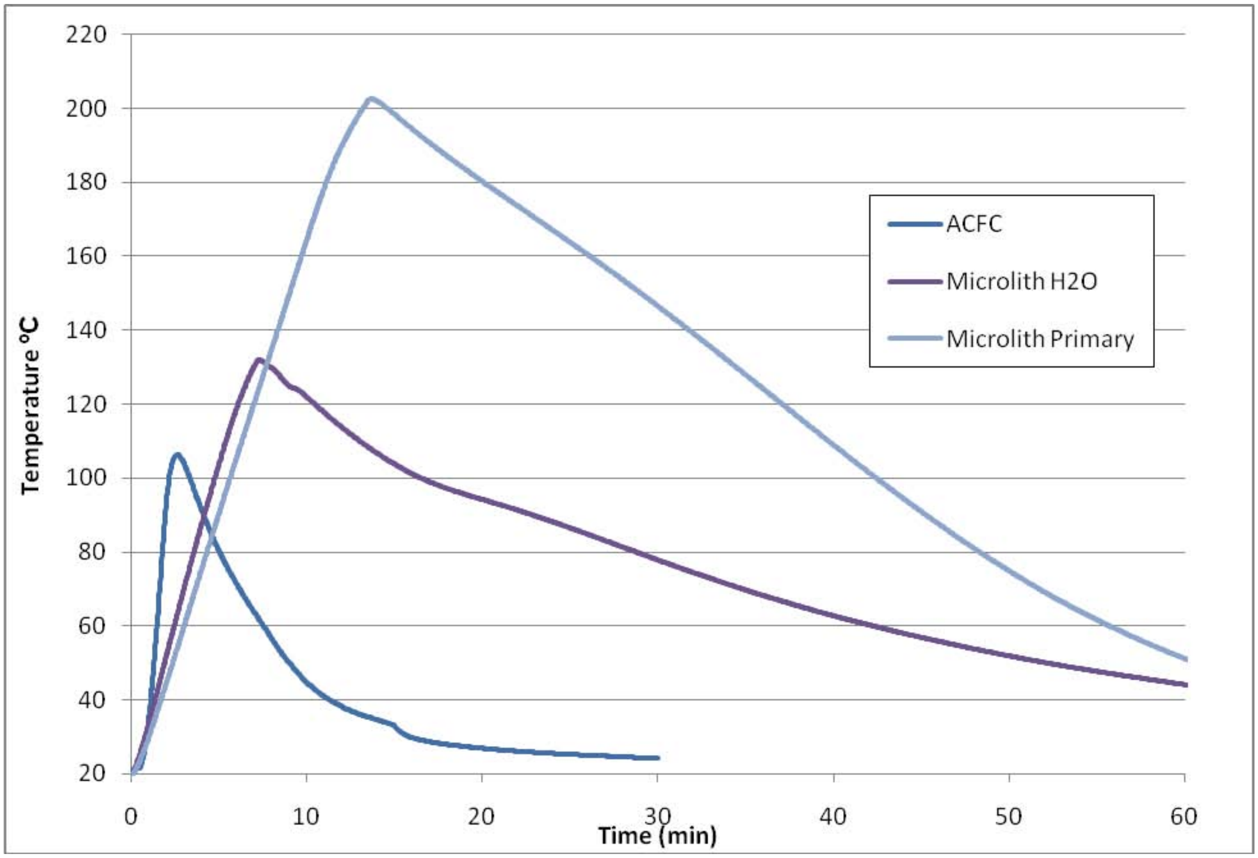

Figure 9: Single Heating and Cooling Cycle for ACFC, Microlith™ Water-Removal Module, and Microlith™ Contaminant Removal Module

Cyclic filter testing was performed as much as the filters permitted. The CFCMS filters were not tested cyclically due to the aforementioned shorting. One set of the Microlith™ filters also experienced electrical shorting, so a single set was used. Water adsorption at 50% RH (Figure 10) was demonstrated with almost complete removal at only 34W/CFM. Although total water removal requires a significant amount of energy, this eliminates the latent heat load from an environmental control unit (air conditioner) operated in conjunction with the filter system, and a portion of this load may be considered an energy offset. The Microlith™ also demonstrated total capture of a pulse of R-134a at 10,000 mg/m^3 for 60 seconds, and complete desorption in 20 minutes.

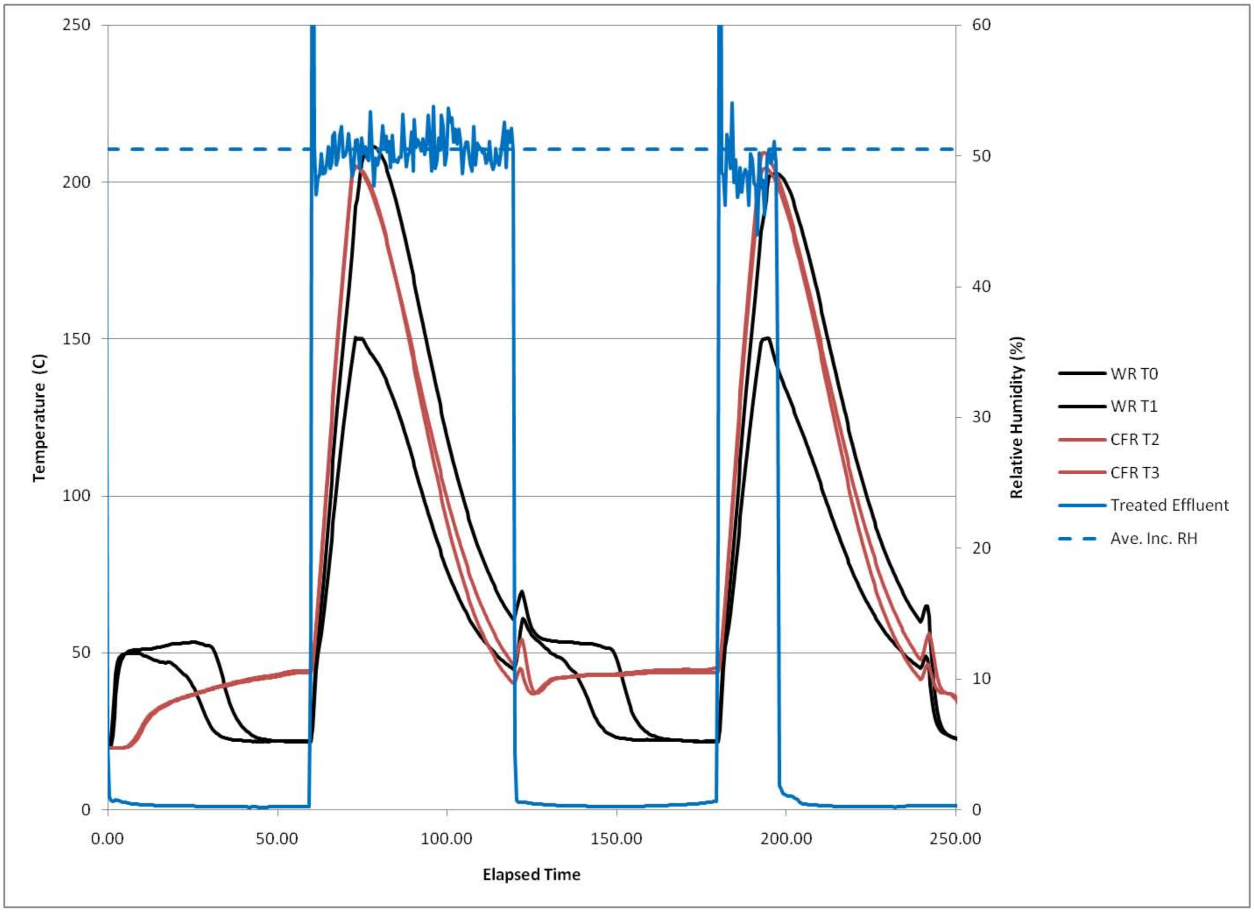

Figure 10: Water Adsorption with a Microlith™ Water-Removal Module and Microlith™ Contaminant Removal Module in Series. Humid Air is Bypassed During the Regeneration Cycle

Cyclic heating and cooling of the ACFC system in both dry air and 50% humidity (Figure 11) demonstrates that half-cycle times as low as 15 minutes can be achieved. The power input was the same for all cycles, so the difference in the peak temperatures is the result of the humidity in the second half of the test. Power consumption was 16W/CFM. The effluent humidity remained around 50%, with only a small portion of the water vapor being adsorbed. By incorporating some layers of hydrophobic ACFCs in the design, some pores will remain available to adsorb contaminants even in high-humidity conditions.

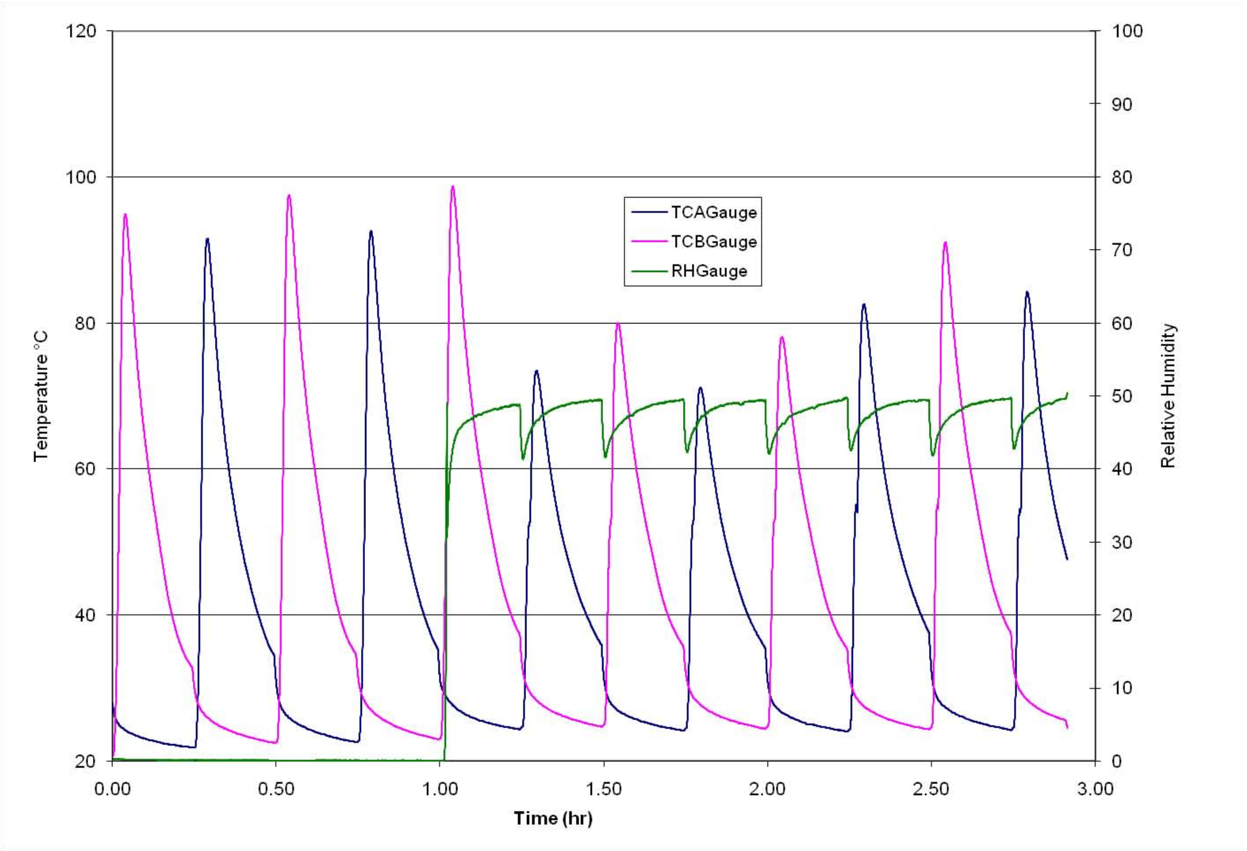

Figure 11: Cyclic Operation of ACFC Adsorbers in Dry Air and with 50% RH at Constant Power.

Continuous capture of propane was demonstrated (Figure 12) with an influent concentration of ~2200 ppmv. Although there is some propane penetrating the system, this is an artifact of the channeling/leakage identified in the breakthrough curves (Figure 8) and not reflective of the potential performance of the system if this mechanical leakage is eliminated.

Chemical treatments of ACFCs (Mangun, et.al. 1999), (Mangun, et.al. 2001) demonstrate that both acid gases and base-forming gases (TICs) can be captured at reasonable efficiencies. Isotherms for both ammonia and sulfur dioxide are comparable to those of propane onto untreated ACFC with similar challenge concentrations (1000-5000 ppmv). With chemically-treated ACFCs, nearly all TICs can be rejected by an ESA system, while meeting DTRA objectives for cube, removal efficiency, pressure drop, and power consumption.

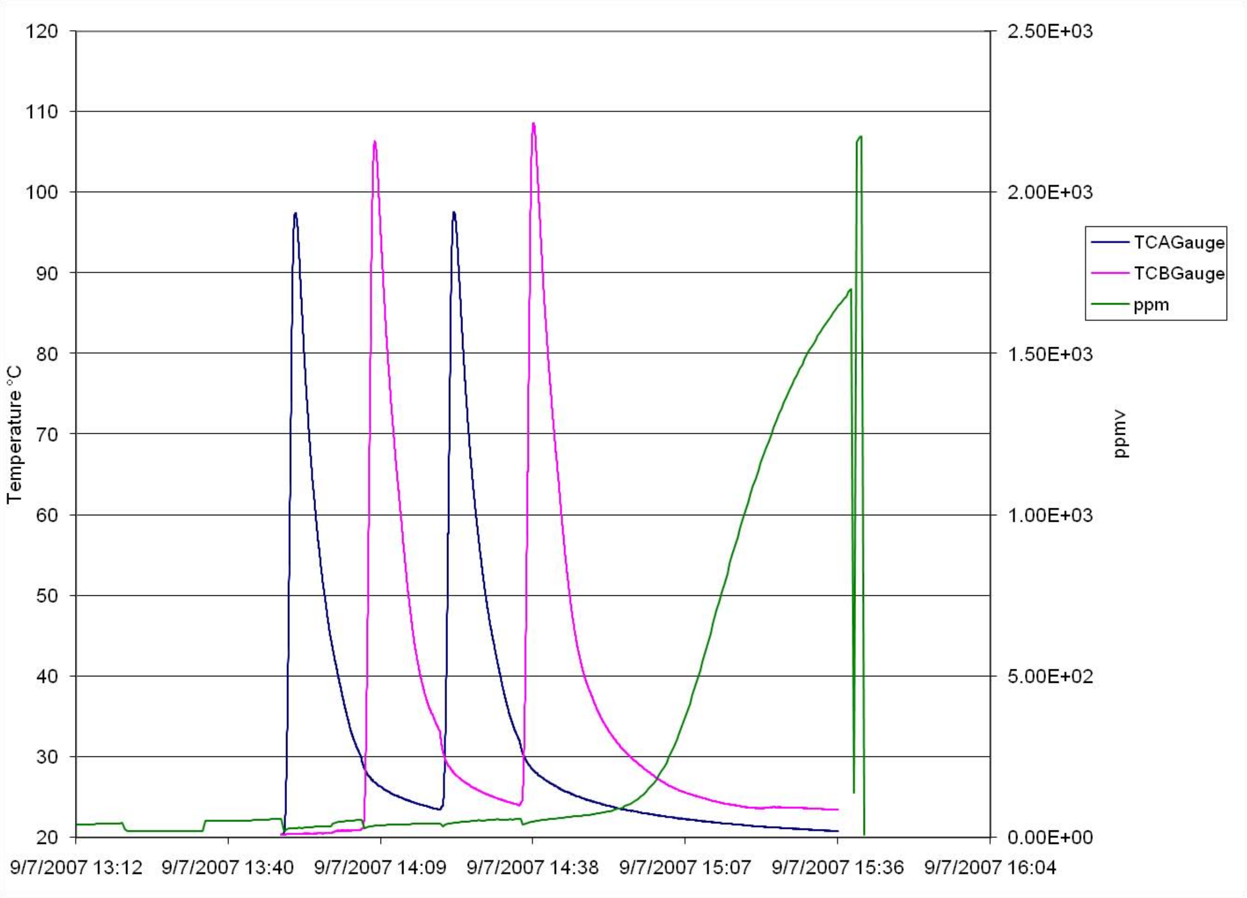

Figure 12: Cyclic Operation of ACFC Adsorbers with a Continuous Challenge of ~2200 ppmv of Propane. Filters Show Potential to Reject an Infinite Challenge if the Channeling Can Be Eliminated

CONCLUSIONS

The three ESA filter media (ACFC, CFCMS, and Microlith™) and their corresponding filter morphologies were evaluated to assess the potential for ESA as a practical regenerative filter technology for collective protection. Although each of the entrants experienced difficulties in their first iteration, such as channeling, poor bed utilization, and electrical shorting, the technology appears to be a viable option. The cube of the systems was 2-3 times the DTRA objective of 0.15 CF/CFM, but with further optimization and scaling, the goal is achievable. Power consumption was within the DTRA metric of 50W/CFM for both the Microlith™ and ACFC, with the Microlith™ providing a significant energy offset to an associated environmental control unit during warm weather. The ACFC units demonstrated sustained protection, under the energy budget, with low pressure-drop, at a challenge level equivalent to ~10,000mg/m^3.

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