

RECIRCULATING THERMOCATALYTIC AIR PURIFIER FOR COLLECTIVE PROTECTION

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ABSTRACT

MesoSystems Technology Inc. is developing a recirculating air purifier that deactivates chemical and biological contaminants without the need for replaceable filters or adsorbents. The thermocatalytic air purifier (TCAP) is based on three enabling technologies: (1) lightweight, high-effectiveness heat exchangers; (2) aerogel-based insulation; and (3) Honeywell's Military Air Purification catalyst, which is proven capable of oxidizing a wide variety of chemical agents.

TCAP prototypes have demonstrated high single-pass destruction efficiencies for simulated chemical and biological agents. This paper describes the development history of the TCAP, the enabling technologies, and the design tradeoffs that were considered.

1.0 INTRODUCTION

Thermocatalytic air purification involves contacting potentially contaminated air with a suitable catalyst at high temperatures (typically 250°C to 400°C). Chemical agents are deactivated by oxidation and biological agents are rendered ineffective by the high-temperature exposure.

The U.S. Department of Defense has funded the development and testing of catalysts for deactivation of chemical agents. Catalysts have been developed that provide excellent destruction of a wide variety of chemical agents.¹⁻⁷ However, thermocatalytic air purification has not yet displaced any of the adsorbent-based air purifiers in the U.S. military. Adoption of thermocatalytic air purification has been slowed by two principal challenges: 1) acid-gas generation; and 2) power requirements for heating and cooling the air.

This paper describes progress toward resolving the second of these two challenges. MesoSystems Technology, Inc. has developed thin-walled, compact heat exchangers that significantly decrease the energy requirements of thermocatalytic air purification. These heat exchangers are integrated with a suitable oxidation catalyst to create a recirculating air purifier.

2.0 BACKGROUND

This section provides a brief overview of thermocatalytic air purification, its advantages and disadvantages, and some of the design tradeoffs that were considered in the development of the recirculating thermocatalytic air purifiers (TCAPs) described later in this paper.

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2.1 Basis of TCAP Operation

The thermocatalytic process purifies air without using conventional filter or adsorbent media. Thus, maintenance requirements are reduced because there are no components that require periodic replacement. Instead of filters, heat is used to kill airborne biological organisms and to oxidize the hazardous chemicals to primarily CO₂ and H₂O. Target chemicals include chemical agents such as GB (Sarin) and HD (distilled mustard).

Figure 1 shows the basis of the thermocatalytic air purifier design. A high-effectiveness heat exchanger is coupled with a heated reactor, which contains an appropriate oxidation catalyst. Unpurified air enters the heat exchanger and is heated by the purified air exiting the reactor. The incoming air enters the reactor area and contacts the catalyst, which induces the oxidation of a wide variety of organics to carbon dioxide and water.

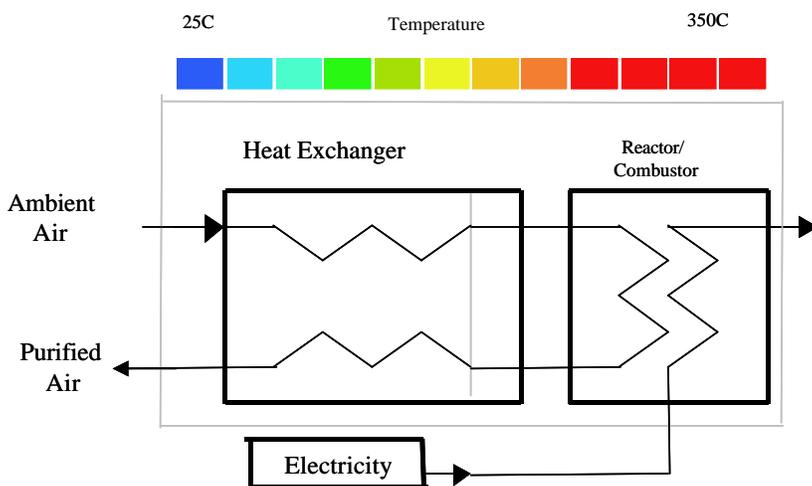


Figure 1. Flow diagram for TCAP operation.

Non-combustible portions of chemical-agent molecules either deposit onto the catalyst as an acid (e.g., phosphoric acid) or are released as acid-gases (e.g., NO/NO₂). The deposited acids accumulate and will eventually degrade catalyst performance. For the envisioned collective-protection application, however, the threat concentrations are low enough (*ca.* 0.03 mg/m³) that very little degradation is expected even with long-term operation. Released acid gases are also not cause for concern because of the low concentrations involved.

2.2 Advantages of Thermocatalytic Air Purification

Thermocatalytic air purification offers several advantages over carbon-based systems. Some of these advantages are:

Performance: Thermocatalytic air purification can effectively destroy a wide variety of chemical and biological agents. Destruction efficiencies of greater than 99.9999% can be achieved. Carbon-based systems can remove (not necessarily destroy) many chemical agents, but not biological agents. HEPA filters are often included in carbon-based systems to provide protection against biological agents. Further, some toxic chemicals are not adequately captured by carbon adsorbents, so carbon-based systems will not provide protection against these chemicals. The decontamination efficiency of carbon-based systems can

be degraded by increased temperature and humidity, and some compounds can displace adsorbed chemical agent thereby reducing the adsorbent effectiveness.

Lifetime: Carbon adsorbents have a limited capacity for some chemical agents. Thermocatalytic air purification, however, provides protection against a large number of attacks. The principal mechanism of performance loss is catalyst deactivation due to adsorption of phosphoric acid (from organophosphorus agents such as GB) onto the active catalyst sites. The life expectancy of the catalyst is, therefore, dependent on the total amount of phosphorus-bearing agent oxidized. Based on an assumed GD threat concentration of 0.029 mg/m³ for the recirculating filter air purifier, no significant catalyst deactivation is expected for at least 12 years of continuous service.

Storage: Carbon-based adsorbents must be properly stored otherwise they can become less effective over time as the adsorption sites become saturated with water and/or semi-volatile organics. The performance of thermocatalytic systems does not significantly degrade during storage.

Logistics: Carbon adsorbents must be periodically replaced and disposed. Spent adsorbent must be treated as hazardous waste if it has been exposed to chemical agents. Supplying new adsorbent and collecting spent adsorbent can constitute a significant logistical difficulty. Thermocatalytic systems require infrequent maintenance, so the logistical burden is greatly diminished. The thermocatalytic system, however, may require some additional logistics support to account for its likely greater mass and higher electric-power requirement (or combustion fuel requirement).

2.3 Recirculating TCAP Design Considerations

There exists a military need for air purification inside chemical/biological collective-protection shelters. These shelters are maintained under a slight positive pressure using purified air from adsorbent-based purification systems. Despite the positive pressure, some chemical and biological agents are expected to be present in the air inside the shelters. These agents are carried into the shelters via several mechanisms (e.g., contaminated personnel entering the shelter). Currently, the U.S. Military has fielded the Recirculating Filter Unit (RFU), which is a carbon-adsorbent-based device designed to adsorb GB and HD agents at low concentration levels. The RFU is shown in Figure 2.



Figure 2. Military RFU

The RFU has a limited capacity for collection of chemical agents and it is unlikely to effectively remove biological agents. With these limitations in mind, the Defense Sciences Office of the Defense Advanced Research Projects Agency (DARPA) funded MesoSystems Technology, Inc., to develop a recirculating air purifier based on thermocatalytic oxidation. Air purification systems based on thermocatalytic oxidation have previously been developed and demonstrated, but these systems were thermally inefficient and generated undesirable levels of acid gases at high threat concentrations. High-effectiveness, thin-walled heat-exchangers make possible a highly efficient thermocatalytic oxidation system, and the low threat concentration inside collective-protection shelters eliminates concerns over acid gases.

Engineers at AlliedSignal Corp. (now Honeywell) previously have demonstrated a recirculating air purifier based on a low-temperature oxidation catalyst coupled with a recuperative heat exchanger.⁸ High conversion rates were observed for a variety of chemical contaminants, but the heat exchanger used for this effort had a lower

effectiveness (about 0.70) than the heat exchangers used for the prototypes described in this paper.

MesoSystems' DARPA-funded air purification efforts are directed principally at developing a recirculating air purifier. This device is designed to recirculate and purify air inside a room or other collective-protection shelter (e.g., field hospitals for chem-bio protection). The performance goals for the MesoSystems TCAP are:

- Flow Rate > 0.2 m³/s (350 CFM)
- Purification Efficiency > 90%
- Required Power < 2 kW
- Volume < 110 L (4 ft³)
- Mass < 30 kg (66 lbs)

Prototype TCAPs have been tested against a range of simulated chemical and biological contaminants. Simulated anthrax spores (i.e., *Bacillus globigii*) are rapidly destroyed by the thermocatalytic air purifier.

Three recently developed technologies make the TCAP possible. These are: 1) High-effectiveness, thin-walled heat exchanger/reactors developed by MesoSystems; 2) Ultra-low-thermal-conductivity, aerogel-based, high-temperature insulating panels developed by Nanopore, Inc. (Albuquerque, New Mexico); and 3) Military Air Purification catalyst from Honeywell (Des Plaines, Illinois), which is capable of oxidizing a variety of nerve agents. Each of these technologies is discussed below.

2.3.1 MesoSystems High-Effectiveness Heat-Exchanger/Reactor

Heat-exchanger effectiveness is defined as the actual rate of heat transfer divided by the theoretical maximum rate. A 90% effectiveness heat exchanger, for example, transfers 90% of the available heat in the hot gas to the cold gas. Very high-effectiveness heat exchange is an essential enabling technology for thermocatalytic air purification.

Without any heat exchange between the incoming and outgoing air streams, the power required to run the reactor is prohibitively high. For example, a heat-based air purifier without a heat exchanger operating at an air flow rate of 200 CFM requires over 30,000 watts of energy to heat the gas. Further, this heat must then be removed before the purified air can be used.

In contrast, the TCAP operates on about 2,000 watts – slightly more power than a household hair dryer – and efforts are being made to reduce this power requirement further. The high efficiency heat exchanger translates not only into reduced energy costs, but reduced air-cooling costs in warm climates. Because the purified air exits the device at nearly the same temperature as the in-coming dirty air, the additional burden on building air-conditioning systems is small.

Figure 3 shows the relationship between required heat exchanger effectiveness, reactor temperature, and air flow rate for an assumed heater power of 2.2 kW. Increasing the heat-exchanger effectiveness increases the reactor temperature. For the catalyst used in the TCAP, a minimum reactor temperature of about 250°C is required. To achieve the target flow rate of at least 350 CFM, the effectiveness must be at least 0.95.

Through DARPA support, MesoSystems has developed thin-foil and thin-wall-ceramic heat-exchanger designs that are capable of achieving an effectiveness in excess of 95%. The thin channel walls (whether they are composed of stainless steel or ceramic) are required to achieve high effectiveness in a compact, lightweight heat exchanger.

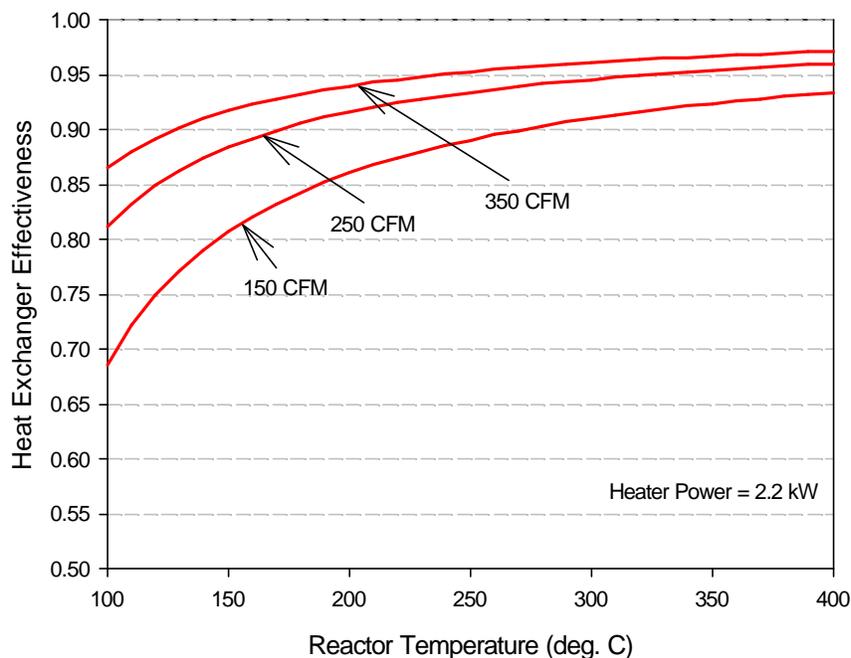


Figure 3. Heat exchanger effectiveness vs. reactor temperature.

The channel walls in Mesosystems' thin-metal-foil heat exchangers are on the order of 25 microns thick, which is equal to the thickness of household aluminum foil. Conventional plate heat exchangers have channel walls at least twenty times thicker (i.e., 0.5 mm). Thicker channel walls result in greater mass than might be expected because of the effect of conductive heat transfer along the channel walls in the direction of air flow. This effect, which is known as longitudinal conduction, can usually be ignored for low-effectiveness heat exchangers, but it often drives the design of high-effectiveness heat exchangers. This is illustrated in Figure 4, which shows how increasing the channel wall thickness in a high-effectiveness heat exchanger results in very large increases in heat exchanger volume and mass. Based on Figure 4, it is clear that the heat exchanger channel walls must be very thin if the TCAP is to be similar in size and mass to the RFU (shown by the horizontal lines). Because ceramic has a lower thermal conductivity and lower density, thicker walls are permissible. A 100-micron ceramic wall thickness is roughly comparable to a 25-micron stainless steel wall thickness in terms of overall system mass and volume.

The calculations used to generate Figure 4 assume a pressure drop through the heat exchanger of 250 Pa (1 in. H₂O). Increasing the allowable pressure drop results in reduced size and mass, but it also results in increased power consumption by the blower that is used to move air through the TCAP. A pressure drop of 250 Pa was selected as the maximum allowable for the TCAP because larger pressure drops result in unacceptably large blower power consumption. At 250 Pa and 350 CFM, the blower consumes roughly 200 W, which is 10% of the heater power. Most of the blower power cannot be recovered in the heat exchanger, so the blower power must be kept low.

In addition to the thin-walled heat-exchanger technology, MesoSystems developed a novel, integrated reactor/heat-exchanger design specifically for the Thermocatalytic Air Purifier. This patent-pending design eliminates the need for manifolds and sealants in the heated regions of the device. Not only does this design result in a much smaller unit, it also eliminates the problem of leak-induced catalyst bypass, which can significantly reduce the unit's effectiveness.

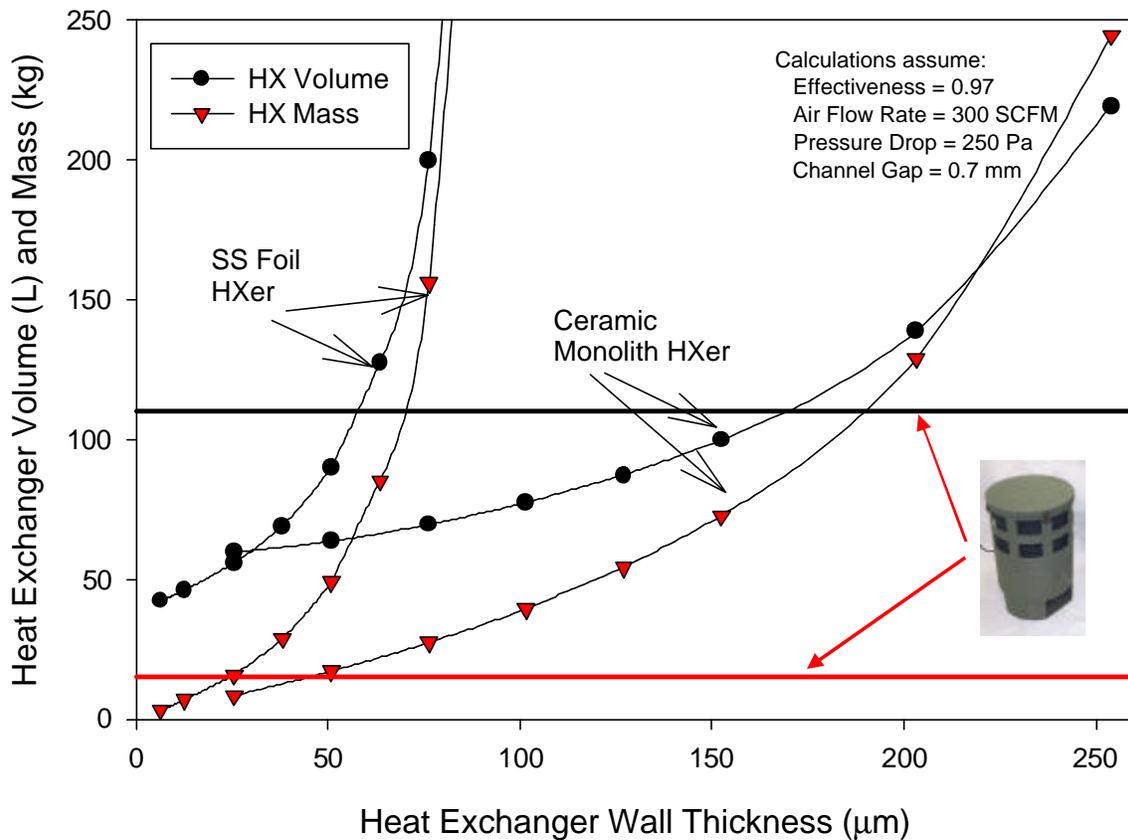


Figure 4. Heat exchanger size and mass versus wall thickness.

2.3.2 Nanopore Vacuum Insulation Panels

With support from DARPA via a MesoSystems subcontract, Nanopore, Inc. (Albuquerque, New Mexico) has developed aerogel-based, vacuum insulation panels suitable for use at the high temperatures (up to 400°C) inside the TCAP. Nanopore currently sells low-temperature vacuum insulation panels to the refrigeration and shipping industries. Nanopore developed a Kapton-coating technique that allows the aerogel panels to maintain vacuum at temperatures up to 400°C.

The aerogel-based insulation is needed for the TCAP to keep heat losses to ambient at reasonable levels. Typical high-temperature insulating materials have thermal conductivities in the range of 0.05 to 0.1 W/mK. The thermal conductivity of NanoPore’s insulation, however, is about 0.005 to 0.01 W/mK at high temperatures – roughly a factor of ten lower than conventional insulation. This high-performance insulation allows the TCAP to be more energy efficient (because less heat is lost to ambient) and considerably smaller than if conventional insulation was used. To keep heat losses to manageable levels (i.e., less than about 50 watts), 1.5-cm-thick aerogel panels are used to surround the TCAP. If conventional insulation was used instead, the insulating layer would need to be about 15 cm (6 inches) thick, thereby increasing the overall volume of the device by a factor of 2.5. The estimated heat loss from the TCAP for the two insulation types is shown in Figure 5.

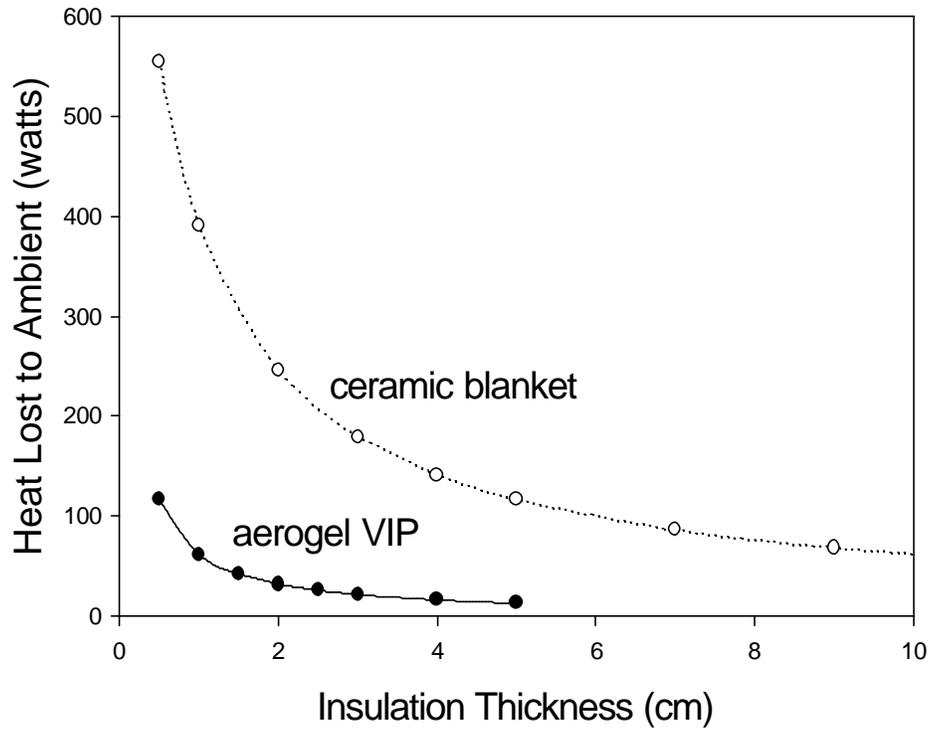


Figure 5. Heat loss from TCAP vs. insulation thickness and type.

2.3.3 Honeywell’s Military Air Purification Catalyst

The Military Air Purification (MAP) catalyst resulted from catalyst-development work conducted at AlliedSignal Corp. and SBCCOM over the past two decades. The catalyst composition and deposition method are proprietary to Honeywell, but Honeywell has agreed to supply MAP catalyst to support development of the TCAP. The MAP catalyst was selected for use in the TCAP because it has been tested and proved effective against a wide variety of chemical agents including GB, GD, HD, VX, AC, CG, CK, and L. Because live-agent testing is expensive, we decided to use a catalyst that has already been proven effective.

3.0 PROTOTYPE DEVELOPMENT AND TESTING

Initially, our project goal was to develop a very compact, lightweight heat-exchanger/reactor that could purify up to 100 LPM of air using a thermocatalytic approach. This device could, in effect, function as a gas mask. Thin-foil heat exchangers were developed using 25-micron-thick stainless steel foils. We began work on the TCAP as an extension of the thermocatalytic gas mask work. For this reason, our early TCAP prototypes were constructed using 25-micron stainless steel foils. Figure 6 shows the conceptual arrangement of the foil, catalyst, and air-supply headers.

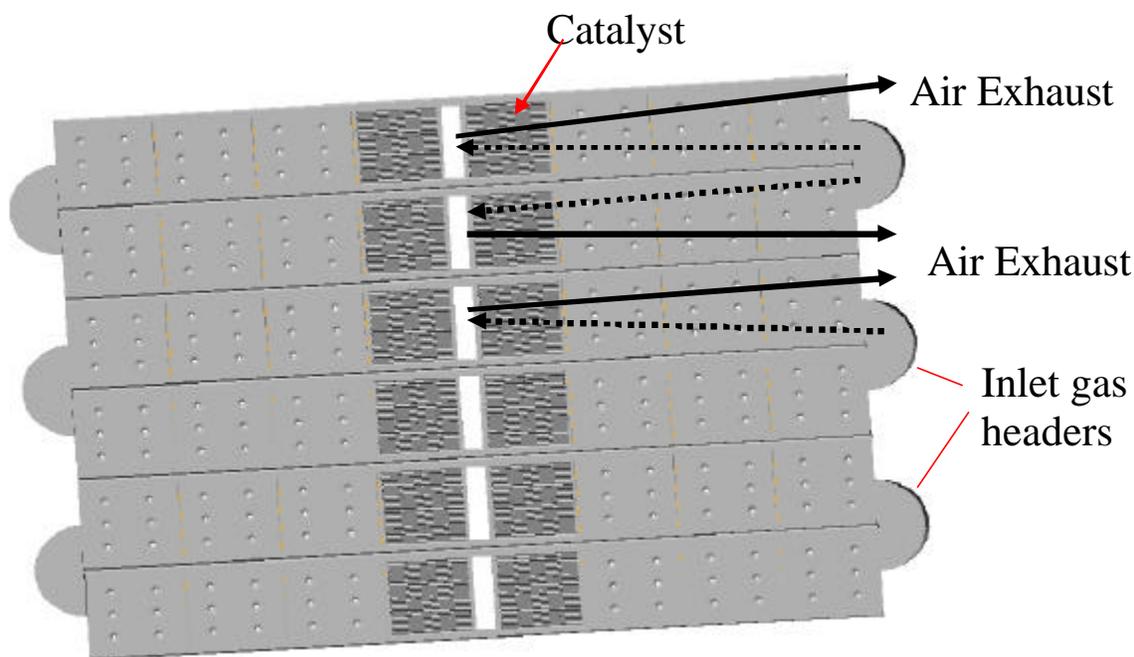


Figure 6. Flow Direction of Air through the Thin-Foil TCAP

The foil shown in Figure 6 is approximately 30 cm wide and 45 cm long. Foil sheets are stacked on top of each other and sealed along the edges by metal gaskets. Prototypes with up to 650 foil sheets have been constructed. The air-delivery manifolds are designed such that each channel (formed between successive foils in the stack) is either for air flowing in or for air flowing out. Further, the “air in” alternate with the “air out” channels so each “air in” channel, for example, is sandwiched between two “air out” channels. This arrangement creates the counter-current heat exchange that is required to achieve high heat-exchanger effectiveness.

Computational modeling and testing revealed the optimum channel gap is approximately 0.7 to 0.8 mm (when using relatively flat channel walls and 25-micron-thick foils). This channel dimension yields the smallest and lightest possible heat exchanger for this application. The channel gap is maintained by a combination of 0.75-mm-high dimples and surface features in the foil that impart rigidity.

As shown in Figure 6, air flows into the heat-exchanger stack through six inlet-gas headers (shown as the semi-circles on the left and right sides of the foil). Air enters each “air in” channel and flows toward the central region, which contains both the catalyst and the electric heating elements (see dashed arrows in Figure 6). The heating elements are inserted through the rectangular openings in the center of the foil stack. After passing over the catalyst, the air flows up or down through the rectangular openings before flowing back out of the heat exchanger via the “air out” channels (see solid arrows in Figure 6). The “air in” and “air out” channels are determined by placement of thin spacers near the entrance/exit region of each channel.

Figure 7 shows a prototype formed from 250 foil layers and a second prototype formed from 650 layers. These TCAPs were tested against a variety of chemicals to determine their single-pass destruction efficiencies and clean-air delivery rates (CADR).



Figure 7. 250-layer TCAP (left), RFU (center), 650-layer TCAP (right)

The CADR of an air purifier can be determined either by directly measuring the single-pass removal/destruction efficiency and the flow rate or by performing a concentration-decay test. In a concentration-decay test, the air purifier is operated inside a sealed enclosure of known volume. At time = 0, a known quantity of challenge agent (or simulant) is injected into the enclosure. The chamber must be well-mixed, so additional fans are often used to provide agitation of the air. The concentration of the challenge agent decays exponentially (assuming constant flow rate and destruction/removal efficiency) according to the equation:

$$C(t) = C_o \exp\left(-\frac{hQt}{V}\right)$$

where C = challenge agent concentration
 C_o = initial concentration
 h = single-pass destruction/removal efficiency
 Q = air flow rate through the purifier
 t = time
 V = volume of test chamber

The data from a concentration-decay test can be fit to an exponential-decay equation to determine the values of C_o and hQ . The CADR is equal to hQ .

3.1 Chemical Destruction by TCAP Prototypes

The concentration-decay approach was used for the chemical-destruction tests at MesoSystems. Figure 8 shows example test data collected using the 650-layer TCAP inside a 14.5 m³ enclosure. Acetone was periodically injected into the enclosure and the acetone concentration monitored via gas chromatography. Also shown in Figure 8 is the performance of the RFU against approximately the same level of acetone challenge. This comparison illustrates one of the advantages of thermocatalytic air purification: high capacity compared with adsorption-based approaches.

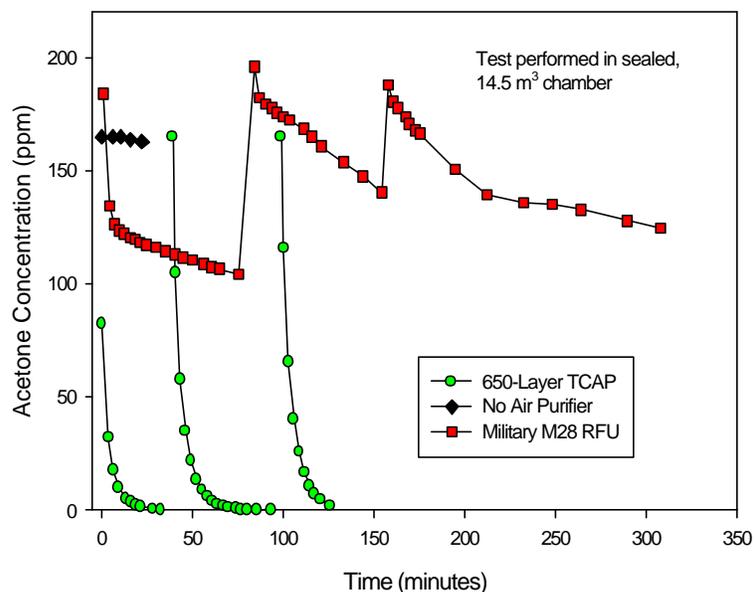


Figure 8. Concentration decay profiles for 650-layer TCAP and the RFU for acetone.

Table 1 below summarizes the CADRs obtained when operating the 650-layer TCAP at a total air flow rate of 170 cfm.

Table 1. Destruction of chemicals by the 650-layer foil TCAP

Chemical	CA DR (cfm)	Single-Pass Destruction Efficiency (%)
Propane	62	36
Hexane	88	52
Acetone	94	55
Methyl acetylene-propadiene	112	66
Hydrogen	140	82

The destruction rates vary considerably depending on the chemical used as the challenge agent. Catalytic destruction susceptibility varies depending on the types of bonds present in the challenge molecule. Short-chain hydrocarbons are usually more resistant to oxidation than are larger molecules or molecules with other functional groups present (e.g., aldehydes and ketones).

The relevant question, of course, is the oxidation resistance of the live chemical agents. Considering their molecular structure, it seems likely that chemical agents will tend to exhibit greater susceptibility to thermocatalytic oxidation.

Tests of the 650-layer prototype were conducted at the Edgewood Chemical and Biological Center (ECBC; Aberdeen Proving Ground, Maryland) using simulated chemical agents. Dimethyl-methyl phosphonate (DMMP) was used to simulate GB/GD agents, and chloro-ethyl-ethyl sulfide (CEES) was used to simulate HD agent. The TCAP's large surface area of stainless steel was found to very effectively adsorb the DMMP at the challenge concentration of 10 mg/m³, and >95% removal rates were observed for several hours regardless of whether the TCAP heater was activated.

The CEES tests were also conducted at 10 mg/m^3 . At steady state, the CEES destruction efficiency was determined to be 70 to 80%, which implies a CADR of 140 CFM for CEES using this prototype. The 650-layer foil TCAP has an overall mass of 55 kg and a heat-exchanger core volume of 100 liters. Electric heaters provide heat to the reactor at a power of 2.2 kW. A centrifugal-fan blower provides 200 CFM of air flow at a power consumption of 0.2 kW. The air-purification power efficiency of this prototype for CEES is, therefore, about $(140 \text{ CFM})/(2.4 \text{ kW}) = 58 \text{ CFM/kW}$.

Heat exchanger effectiveness of the 650-layer foil prototype was limited to about 0.93 by a combination of pressure-induced foil flexing and non-uniform flow distribution created by the centrifugal blower and the air-delivery manifolds.

In an effort to overcome the problem of foil flexure, we switched our focus from foil-based heat exchangers to thin-walled ceramic heat exchangers. Ceramic has the advantages of increased rigidity, lower bulk density, and lower thermal conductivity than stainless steel. As mentioned earlier, lower thermal conductivity is often desirable for high-effectiveness heat exchangers to reduce the deleterious effect of longitudinal conduction. Figure 4 showed that ceramic-based heat exchangers can be made with thicker walls (roughly 4 times thicker) than stainless-steel heat exchangers with no change in performance, size, or mass.

Figure 9 shows a 90-CFM TCAP made using thin-walled ceramic heat exchangers. Extruded cordierite ceramic monoliths are modified to create the heat-exchanger/reactor components. These monoliths are produced as catalyst substrates and are widely used in automobile catalytic converters. The 90-CFM ceramic TCAP is composed of 30 blocks of cordierite ceramic 10 cm wide, 5 cm high, and 15 cm long. The ceramic wall thickness is 100 microns (0.004 inches) and the ceramic channels are square with a dimension of 1 mm. The ceramic monolith blocks are modified in such a way as to create a counterflow heat exchanger with integral reactor section – the resulting flow path is similar to that shown in Figure 6. The exact process by which this is accomplished is considered proprietary at this time. Multiple blocks are configured together such that they process air in parallel. An electric heater is also built into the hot end of each block.



Figure 10. Ceramic TCAP prototype.

The TCAP shown in Figure 9 is composed of 30 ceramic blocks. Each block is coated with Military Air Purification catalyst (from Honeywell). This device is roughly cubical with each side measuring 35 cm. Its mass is 27 kg of which 13 kg is the heat-exchanger/reactor core. The heater consumes 0.9 kW of electrical energy and the blower requires 0.1 kW at full speed. Heat exchanger effectiveness is 0.95.

This unit has been tested against a variety of chemicals including propane, acetone, hexane, acetylene, and CEES. The air-purification power efficiency of this device is somewhat improved compared with the foil prototype. For CEES, the efficiency is about 70 CFM/kW. We have made some recent modifications to the heat exchanger designs that in single-block tests have yielded efficiencies as high as 150 CFM/kW. A larger-scale unit has been built employing these modifications and it is currently undergoing tests to evaluate its performance.

3.2 Destruction of Simulated Biological Agents by the TCAP

The 90-CFM ceramic prototype was also used for tests aimed at determining the TCAP effectiveness for deactivation of biological agents. *Bacillus anthracis* (anthrax) spores are generally believed to be one of the most challenging biological agents for heat-based air purification methods.⁷ Anthrax spores are often simulated using *Bacillus globigii* (*Bg*) spores. We supplied the ceramic TCAP prototype with aerosolized *Bg* spores at an inlet concentration of 50 to 100 colony forming units (CFU) per liter of air. The air discharged from the TCAP was sampled and filtered, and the filters were cultured to reveal the number of viable *Bg* colonies. The tests were run with the TCAP operating at between 80 and 95 CFM, which corresponds to a residence time inside the reactor portion of the TCAP of about 0.06 seconds.

There is some concern that *Bg* spores are less heat resistant than anthrax spores. To address this concern, similar tests were conducted using *Bacillus stearothermophilus* (*Bs*) spores, which are generally accepted to be more heat resistant than anthrax spores. The results for the *Bg* and *Bs* spore-deactivation tests are shown in Figure 11. At the *ca.* 0.06 second residence time used for these tests, it appears a reactor temperature of about 350°C is required to achieve high single-pass deactivation of *Bg* and *Bs* spores. This result is also expected to apply to anthrax spores, because the thermal resistance of anthrax spores is thought to be between that of *Bg* and *Bs* spores. Also shown in Figure 10 are data for the deactivation of *Bg* spores at roughly 5X longer exposure times. In these tests, >99.999% deactivation of spores was observed at 225°C and higher.

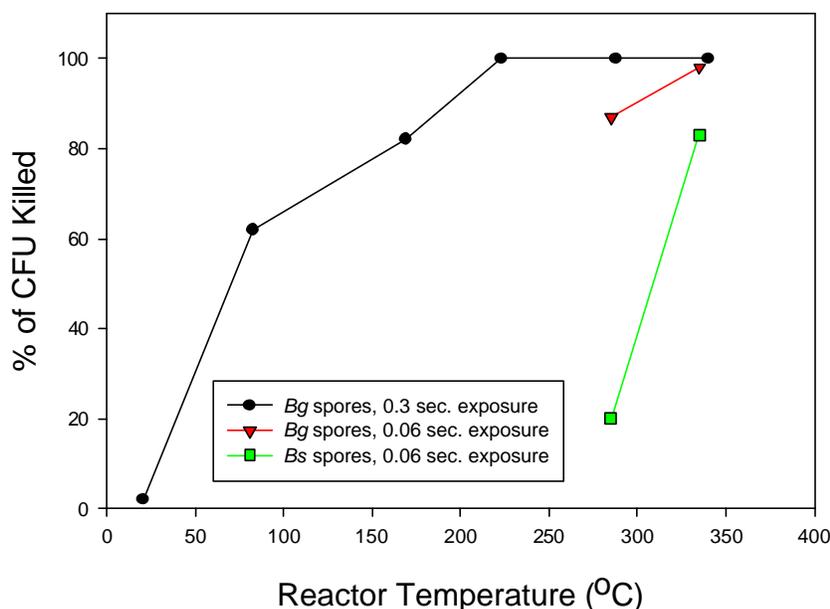


Figure 11. Thermal destruction of *Bg* and *Bs* spores.

The results shown in Figure 11 are reasonably consistent with those of Shankle,⁷ who performed thermal deactivation tests using *Bg* spores in a different reactor geometry. Shankle's data imply complete sterilization of *Bg* spores at a temperature of 345°C for a residence time of 0.06 seconds and 280°C for 0.3 seconds.

4.0 CONCLUSIONS

A combination of high-effectiveness heat exchangers, low-thermal-conductivity insulation, and a suitable oxidation catalyst were combined to create a recirculating air purifier based on thermocatalytic oxidation.

Incorporation of the high-effectiveness heat exchanger is essential for reduction of power consumption by the heater inside the TCAP. We believe thermocatalytic oxidation represents a promising technology for military and commercial air purification because of the reduced logistical burden (i.e., no need for filter replacement) and enhanced protection provided against a wide variety of chemical and biological agents as well as toxic industrial chemicals.

The high-effectiveness, compact TCAP prototypes described in this paper demonstrate that power-consumption issues can be effectively managed through the use of thin-walled heat exchangers and high-efficiency insulating materials.

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