A NOVEL HEPA FILTER ENCAPSULATION PROCESS

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ABSTRACT

Waste management engineers at Lawrence Livermore National Laboratory have developed an innovative process for the treatment of contaminated HEPA filters. The In Situ Stabilization and Filter Encapsulation (IS SAFE™) Process provides several advantages over existing HEPA filter treatment processes. Treatment is accomplished by filling a spent HEPA filter with a low viscosity resin that cures to form a solid monolith. Once a solid monolith has been formed, the HEPA filter has been transformed in two ways: 1) worker hazards/risks associated with handling the filter are eliminated and 2) the in situ encapsulated filter meets LDR requirements and can be disposed of in a permitted landfill. A patent has been filed for this process. The IS SAFE process will be applicable to hazardous, mixed, and low level radioactively contaminated spent filters. Spent HEPA filters are prepared for processing by attaching vacuum fittings to the top and bottom openings of the filter. These fittings are used to attach the filter to a vacuum system comprised of a vacuum pump, vacuum gauge, resin reservoir and associated tubing and fittings. Resin is delivered to the filter in a controlled fashion. The total processing time for a 50-cubic feet per minute (cfm) filter is approximately 25 minutes. The resin filled HEPA filter is allowed to cure at ambient temperature and pressure for at least 24 hours. The final product is a solid monolith with less that 0.1 percent by volume of void space and 100 percent of the filter media coated with resin. Proof of concept studies have been completed using 50, 135, and 1000 cfm closed face HEPA filters. During these studies, we were able to develop and demonstrate a resin delivery process that yielded a final product that was suitable for in ground disposal. Additional adaptations of the process may be required for specific application, but the process equipment, supplies, and methodology have been fully established for contact handled, closed face, contaminated HEPA filters.

INTRODUCTION

High Efficiency Particulate Air (HEPA) filters are used to remove contaminants from air in order to protect worker health and the environment. They are often used in the most hazardous of environments as a last line of defense against a variety of airborne contaminants such as radionuclides, toxic metals, pathogenic bacteria, and viruses. HEPA filters are designed to remove at least 99.97% of airborne particles with diameters greater than or equal to 0.3 μm. When the amount of trapped particles in a HEPA filter cause the pressure drop across the filter to exceed a predetermined limit (varies according to application) the spent filter is removed from service and replaced with a new, clean filter. In most instances, spent HEPA filters are highly contaminated and require treatment before they can be safely and properly disposed.

In the Department of Energy (DOE) complex, HEPA filters are presently stockpiled. The DNFSB (Defense Nuclear Facility Safety Board) recommended on March 8, 2000 that DOE should assess the conditions of "confinement ventilation." This report contained several recommendations that have led the DOE to develop new standards for HEPA filters. Changes incorporated in the new standards include more frequent HEPA change-outs, and establishing a schedule for the removal or replacement of old, existing HEPA filters systems. As a result, LLNL (Lawrence Livermore National Laboratory) now requires replacement of HEPA filters every 10 years for dry systems and every 5 years for wet systems. These changes and an increase in the decontamination and demolition of surplus buildings have resulted in a significant increase in the number of HEPA filters that are in need of treatment prior to disposal.

Several technologies have been developed and used to treat HEPA filters for subsequent disposal. Several processes (Tejima and Fujino, 1994; Uehara, 1993; Fellingham et al, 1985; Oma, 1986; Ziegler and Johnson, 1978) include shredding of the contaminated filter followed by incineration, cementation, or vitrification of the shredded HEPA filter fragments to solidify and bind hazardous particles in spent filters prior to disposal. Chemical leaching with hydrofluoric acid, nitric acid, or sodium hydroxide is used in processes developed by Idaho National Engineering and Environmental Laboratory and the Savanah River Site (Chakravarty and Greene, 1995; Brewer and Murphy, 1994; Jantzen, 1997). While all of the previously mentioned processes have been deployed at a wide range of sites, none were deemed suitable for the disposal of mixed waste HEPA filters at LLNL. The primary deterrents to using the existing technologies were the high level of worker exposure with shredding processes and the large volume of spent leaching solution generated with the chemical leaching processes. Also, with cementation processes, the cement interacts with the aluminum separators in the filters and generated hydrogen gas, thereby creating an explosion hazard. One further deterrent to the use of existing processes was the use of elevated temperatures in several of the processes and our desire to conduct treatment at ambient temperatures.

PROCESS DEVELOPMENT CRITERION

In FY99, the LLNL Environmental Protection Department initiated a project with the objective of developing a simple HEPA filter treatment process that could be quickly implemented. In addition, past experience with HEPA filter treatment and characterization demonstrated that we needed an approach that would promote worker safety and still be able to achieve off-site disposal following treatment. The final product of the process developed had to meet the disposal criteria of either a commercial or Government Owned, Contract Operated (GOCO) land disposal facility. Initial efforts focused on meeting the Nevada Test Site (NTS) waste acceptance and disposal criteria. The NTS accepts only non-hazardous waste that meets both federal and state of origin requirements for non-hazardous classification.

Many of the LLNL waste HEPA filters are considered low level mixed waste because they have both hazardous waste properties and radioactive contamination. In general, these HEPA filters are hazardous due to the characteristic of toxicity, which results from

the often high levels of toxic metals captured by the filters during use. We chose to develop a method that would encapsulate or stabilize contaminant particles directly on the filter fabric without substantial handling or destruction of the HEPA filter structure. The primary rationale for this choice was the desire to immobilize the toxic metals in a manner that allowed the treated filter to pass regulatory leach testing so it can be reclassified as low level waste only and be acceptable for disposal at NTS. This encapsulation method would also allow us to avoid the generation of secondary wastes and the regulatory scrutiny of incineration types of treatment.

In review, HEPA filter encapsulation was selected for development because it was the alternative that meet all of the criteria listed below:

- Eliminate generation of secondary wastes
- Simplifies treatment (i.e., produces the desired result by filling a cavity with plastic)
- Reduces potential exposure to personnel
- Precludes intrusive sampling
- Turns characteristic mixed waste into low-level only wastes

The HEPA filter encapsulation process developed at LLNL is often referred to as the In Situ Stabilization and Filter Encapsulation (IS*SAFE) Process. These terms will be used interchangeably throughout this document. The remainder of this paper discusses the encapsulation process in detail and reviews the findings of laboratory studies that demonstrate the ability of the process to meet regulatory requirements for the reclassification of hazardous to non-hazardous waste.

PROCESS DESCRIPTION

With the IS*SAFE Process, a spent HEPA filter is filled with a low viscosity resin that cures to form a rigid, leach resistant monolith. The resin used is a vendor-supplied, two part product that is easily customized to adjust the pot life (time to initial setting) within the range of 0.5 to 3 hours. The polyurethane resin used is typical of those used in casting. The resin includes polyglycols ($C_nH_{2n}(OH)_2$) terminated with isocyantes (-NCO) (primarily Dicyclohexylmethane 4, 4'-diisocyanate) which is activated by polyether polyols with traces of organomercury carboxylatye. This resin has properties, typical of other polyurethanes. Once mixed, the resin has low viscocity and is easily cast. It also has good adhesion and wetting properties, extremely low shrinkage upon curing, is very hard when cured, and has good chemical resistance.

Figure 1 shows the process equipment used in the IS*SAFE process. Inexpensive plumbing materials are used throughout and when they become too difficult to clean, they are discarded and replaced. The resin is mixed in a process vessel with piping connecting it to a clear, plastic sealing flange attached to the bottom of a HEPA filter. The mixing vessel is made of vinyl ester fiberglass has a recessed dished bottom. This allows its use without anchoring and the dished bottom facilitates easy cleaning. The sealing flanges are clear to allow observation of the level of the resin during its introduction. The top of the filter has a similar flange attached to plumbing that terminates at a vacuum pump.

The vacuum pump provides the driving force to introduce the resin into the HEPA filter during treatment. The direction of resin flow in the filter is from the contaminated side to the clean side. In other words, the vacuum pump is attached to the same side of the filter that the blower would be attached to if the HEPA were still in service. Vacuum was chosen for several reasons. First, controlled use of vacuum (along with some gravity feed) allows for the slow, deliberate flooding of the filter cavity. Vacuum also is inherently safer since there is no chance for the build up of pressure and the potential dispersal radioisotopes in the environment. Vacuum also minimizes the development gas bubbles and serves as a de-gassing mechanism for the resin after mixing. The resin is degassed to ensure that the cavity is completely filled and that all filter material and contaminant particles are thoroughly wetted with the resin, prior to curing. A PVC vent valve and vacuum gage are used to control and monitor the HEPA encapsulation process and the operation is manned until the HEPA cavity is full and the system is cleaned out. Cleaning is performed with only a small amount of solvent capable of dissolving the uncured resin.

The first step in the IS*SAFE process is the transfer of both parts of the resin into the process vessel. The resin was formulated so that both parts are introduced at approximately the same volume. Once all the resin has been poured, the mixer is started and the angular velocity is slowly increased. This prevents splashing and provides for smooth mixing. The agitator is one capable of mixing about 55 gallons of material and is similar to those used to mix paint products. During the mixing, bubbles may appear which will remain in the resin throughout the mixing process. Once mixing is complete (approximately 10 minutes), the vacuum pump is turned on and the vacuum is held constant at 1500 Pascal differential in order to remove as many air bubbles as possible from the resin before introduction into the HEPA filter. After a few minutes of degassing, the feed (PVC ball) valve below the process vessel is opened and the resin is allowed to flow into the HEPA filter cavity. Vacuum is gradually adjusted as the HEPA filter interior is filled. The filling continues until there is visual indication that the HEPA filter is completely full of resin. The time required for complete encapsulation varies depending on the size and capacity of the filter. Encapsulation times for the filters evaluated during this project were 20, 30 and 35 minutes for 35, 135, and 1000 cfm HEPA filters respectively. A 1,000-cfm filter takes approximately 35 gallons of resin to fill the cavity. Once the HEPA filter is filled with resin, the vacuum pump is turned off and the feed valve is closed. The resin in the HEPA filter cavity is allowed to cure, while the process equipment is cleaned. It is important that the equipment be cleaned as soon as possible following treatment to prevent the curing of unused resin in the process tank and lines.

Fig. 1. Process Summary photos showing overall process equipment, mixer used, and HEPA cavity being filled

OBSERVATIONS

We installed 9 type J thermocouples in a standard 1,000-cfm HEPA filter prior to encapsulation (Figure 2). The filter was then encapsulated as described above and allowed to cure while measuring and logging temperature readings at each location every 5 minutes. This data allowed us to determine how hot the system would get and how long it would take for the resin to cure. Figure 2 shows the temperature trends during and following the encapsulation of a 1000 cfm HEPA filter. Within 45 minutes of resin delivery to the filter (approx 10 minutes after the resin is mixed) the temperature in the interior of the HEPA begins to increase. The highest temperature of approximately 405 Kelvin was recorded in the center of the HEPA filter, 5 hours after the introduction of the resin. About 6 hours after the introduction of the resin the temperatures begin to decrease gradually. The time to cool down to room temperature was about 1.5 days. The maximum temperature reached on the exterior of the HEPA filter was not recorded but it remained cool enough to touch.

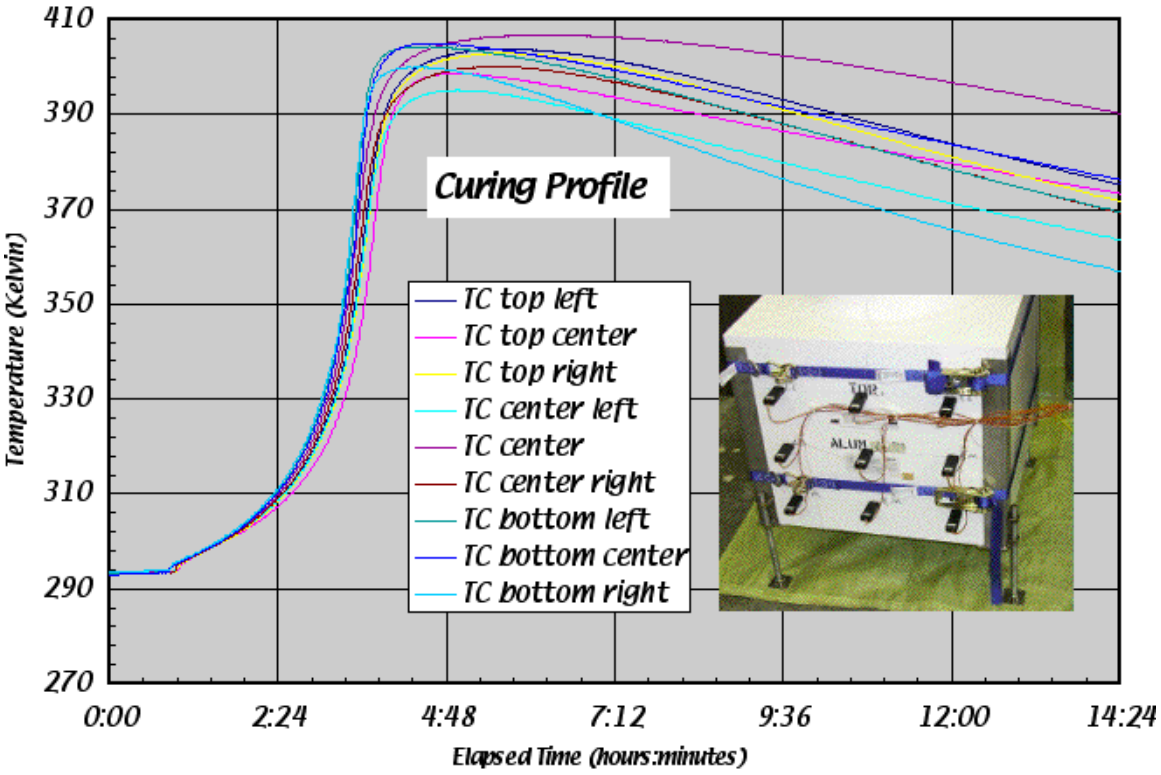

Fig. 2. Temperature profile of the HEPA filter as the resin cures and cools

Three different filter sizes were tested while developing the IS*SAFE process. The first tested were 50-cfm filters, followed by 135-cfm filters, and finally 1,000-cfm filters were encapsulated. It should be noted that all of the development work was completed using new, uncontaminated HEPA filters. Uncontaminated filters were used to minimize worker hazards and allow less restricted handling of the final product. Several of the earlier encapsulation resulted in void spaces in the final product. We determined that the gradual, controlled delivery of the resin to the HEPA is essential to the formation of a void free, solid monolith.

Several HEPA filters have been cross-sectioned using a large throat band saw. The cross sections have been visually examined at several depths from encapsulated filters. All cross sections appear to demonstrate that the resin fully penetrates the filter cavity and completely wets the filter. Filters filled with resin (and cured) were also cross-sectioned after loading the filter with non-hazardous, finely divided particles. This was performed to determine if the resin would fill the cavity uniformly in the presense of particles. We observed that all of the added particles were wetted and encapsulated against the surface of the filter fabric. Figure 3 shows a cross section of a successfully encapsulated filter. The filter is completely penetrated, all the fabric was wetted before it cured, and there are no visible voids in this or any other cross-sections of the HEPA. The apparent void space near the edges in Figure 3 (dark spots or lines) are due to the somewhat imperfect way in which the filter fabric is attached to the wood housing.

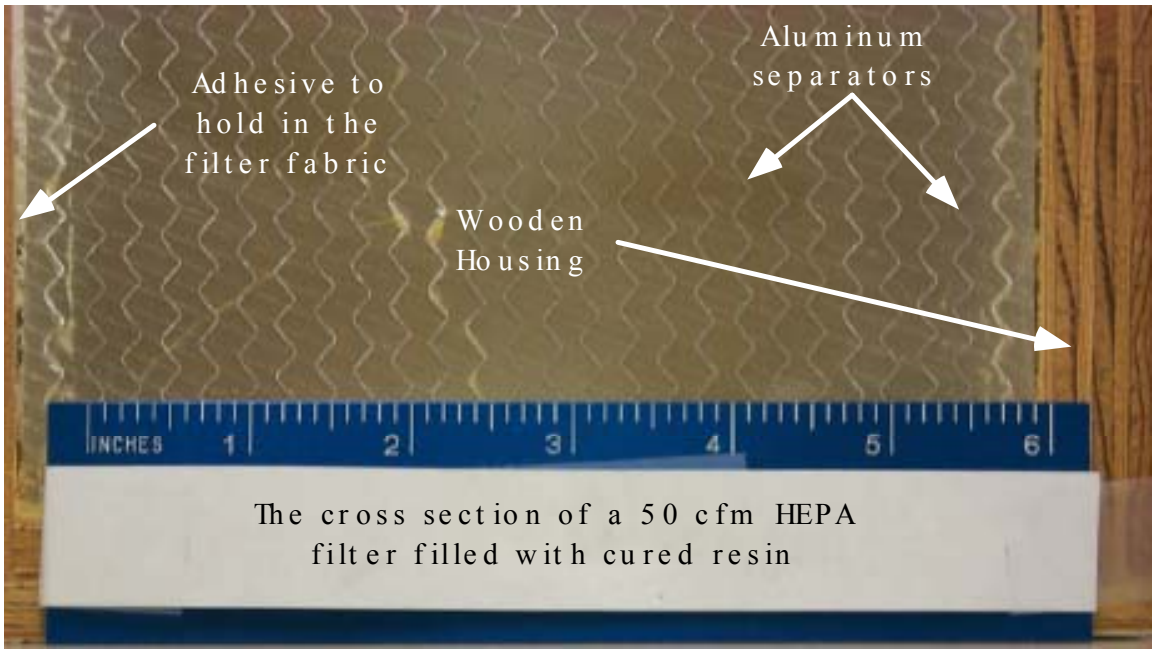

Fig. 3. This photo is indicative of the results when filling the HEPA filter with resin, very few void spaces are found and essentially all material is wetted with resin

Estimates of void space were attempted but they were so small and appeared so infrequently that quantification was not practical (void space is clearly less that a fraction of a percent and occurs rarely and randomly). Bubbles were considered void space and

we did have several that showed up in early experimentation, but with constant monitoring of the feed rate and system vacuum, most of the bubbles would only appear near the surface of the resin well above the filter fabric.

FINAL PRODUCT LEACH TESTING

Mock-up samples were prepared using metal oxides and resin. Oxides were available and are presumed to be the most prevalent species that would be found in the HEPA filter, especially in long term storage in air. These oxides were gravimetrically added to plastic beakers in specified amounts, then premixed resin was added to the beaker so that the resultant mixture of metals and resin was 50 grams. The resin in the beaker was mixed again (by hand) to distribute the metal oxides in the mixture. Since the curing time is long and the viscosity is low, the mixture was occasionally re-mixed to prevent the metal oxides from settling to the bottom. After the sample cured, it was analyzed using the STLC and TTLC procedures. Figure 4 below shows several prepared samples used to determine leaching results of hazardous metals in encapsulated HEPA filters.

Fig. 4. Typical "Hockey Puck" samples prepared as mock-ups for leaching tests

Table I shows the results of the leaching tests using the STLC methodology.

Table I. Results of HEPA filter samples. Data shown are in milligrams per liter in STLC leach effluent

RCRA metal	Compound used	TTLC level	Maximum allowable leachate concentration (mg/L)	Average leachate concentration in test samples (mg/L) (n = 9)	Pass regulatory requirement for non-hazardous?
Chromium	CrO_3	500	5	1.15	Yes
Arsenic	As_2O_3	500	5	0.22	Yes
Cadmium	CdO	100	1	0.21	Yes
Copper	CuO	2500	25	0.21	Yes
Nickel	NiO	2000	20	0.00	Yes
Lead	Pb_3O_4	1000	5	0.10	Yes
Antimony	Sb_2O_2	500	15	0.02	Yes
Selenium	SeO_2	100	1	1.32	No
Zinc	ZnO	5000	250	1.43	Yes
Berylium	BeO	75	0.75	0.00	Yes

With the exception of selenium, all of the metals included in the resin samples were retained sufficiently to allow reclassification as non-hazardous per State of California requirements. Similar test were conducted using the TCLP methodology and all of the metals were sufficiently retained.

CONCLUSIONS

This process will be implemented as a novel way to treat HEPA filters that are mixed waste. We expect to use this process this fiscal year in our existing waste management facilities. Implementation has begun in establishing the required protocol needed to perform this type of treatment in our Nuclear TSD (Treatment Storage and Disposal) Facility. We already have an ISD (Interim Status Document) permit required under the RCRA (Resource Conservation and Recovery Act) and are presently addressing USQs (Unreviewed Safety Questions, a Department of Energy protocol for nuclear facilities). We have approximately 800 filters to manage and this process will provide an opportunity for safe handling and disposal of many of these filters.

This process has some limitations. One is the fact that the final product it is not amenable to size reduction. Any attempt at size reduction after going through this process even with super compaction would be futile. The final product is also heavy and is extremely difficult to sample. Sampling may not be a concern since the Environmental Protection Agency has ruled that debris that has been treated using an approved process, such as microencapsulation, does not require post treatment sampling. The resin used in the development of this process is costs about $100 per gallon. With this resin cost,

treatment of a 1,000-cfm HEPA filter would cost about $4000. Cost may be improved substantially through bulk purchasing of less expensive resin, system automation, and development of process sequences that reduce cleaning and expendable parts needs.

Hazardous waste management engineers at LLNL are eager to implement the process as a means to treat a difficult to manage waste stream that the site is required to dispose of in a timely manner. This process also should be very amenable to high-level waste because it is very simple to implement. Further infrastructure will be needed to determine the best approach in automation and personnel safety. The resin also needs to be evaluated for performance in high radiation fields.

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