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ABSTRACT

Calciner operations and the fuel dissolution process at the Idaho National Engineering and Environmental Laboratory have generated many mixed waste high-efficiency particulate air (HEPA) filters. The HEPA Filter Leach System located at the Idaho Nuclear Technology and Engineering Center lowers radiation contamination levels and reduces cadmium, chromium, and mercury concentrations on spent HEPA filter media to below disposal limits set by the Resource Conservation and Recovery Act (RCRA). The treated HEPA filters are disposed as low-level radioactive waste.

The technical basis for the existing system was established and optimized in initial studies using simulants in 1992. The treatment concept was validated for EPA approval in 1994 by leaching six New Waste Calcining Facility spent HEPA filters. Post-leach filter media sampling results for all six filters showed that both hazardous and radiological constituent levels were reduced so the filters could be disposed of as low-level radioactive waste. Since the validation tests the HEPA Filter Leach System has processed 78 filters in 1997 and 1998.

The Idaho National Engineering and Environmental Laboratory HEPA Filter Leach System is the only mixed waste HEPA treatment system in the DOE complex. This process is of interest to many of the other DOE facilities and commercial companies that have generated mixed waste HEPA filters but currently do not have a treatment option available.

INTRODUCTION

The Idaho Nuclear Technology and Engineering Center (INTEC) located at the Idaho National Engineering and Environmental Laboratory (INEEL) was originally designed to process spent nuclear fuel. In the process, spent nuclear fuel was dissolved and the reusable uranium extracted. The waste is calcined at New Waste Calciner Facility (NWCF) to form a dry powder, and put into concrete storage bins for indefinite storage. NWCF operations involve the handling of fission products, transuranic (TRU) materials, and hazardous materials. High-efficiency particulate air (HEPA) filters are used in the offgas streams to prevent these materials from entering and contaminating the environment. As a result many HEPA filters contaminated with radioactive (5-120 R/hr (β/γ)), transuranic (TRU content >600 nCi/g have been observed), and toxic metal constituents (6-12 mg Cd/g media, 2-4 mg Cr/g media, and 3-6 mg Hg/g media) have been generated (1,2,3). Currently there is no disposal facility that will accept the contaminated HEPA filters as generated. Hence, a HEPA Filter Leach system was designed to lower radiation contamination levels and reduce cadmium, chromium, and mercury concentrations on spent HEPA filter media to allow the treated HEPA filters to be disposed as low-level radioactive waste.

LEACHING THEORY

Leaching is a separation technique that is often used to remove a soluble solid or solute from a solid mixture with the help of a liquid solvent. The HEPA Filter Leach System is primarily concerned with the removal of calcine particles, consisting mostly of metal oxides, that have been trapped by the filter, or salts that have been deposited on the HEPA filter media from entrained liquid in the gas vapor. The trapped particles and salts are removed from the HEPA filter media by contact with a liquid solvent. This process often requires multiple batch leaching cycles or a countercurrent process to adequately remove the contamination from the HEPA filters. Once the contaminants have been removed from the filters excess solvent is removed from the filter media using a couple of filter rinse cycles. The spent solvent and rinse effluent can then be recycled or treated for disposal.

INITIAL PROCESS DESIGN

The HEPA Filter Leach System, located at INTEC in the New Waste Calciner Facility was originally designed to remove the TRU constituents so the filters could be disposed as low-level radioactive waste. The filter leach process was demonstrated in 1983 by leaching two NWCF HEPA prefilters which had been on-line for 81 days. Analysis of these filters showed that the filters were more than 6 times the TRU limit (100 nCi/g). The leaching operation reduced the TRU activity on the filters by 97.9 percent with a single leach (1).

After the HEPA Filter Leach System was designed and constructed in 1988, the Department of Energy decided to impose the Resource Conservation Recovery Act (RCRA) regulations that required the removal of hazardous material from any waste prior to disposal. This changed the operating requirements for the HEPA Filter Leach System to decontaminate the filters for disposal as non-hazardous low-level radioactive waste (3).

TECHNICAL BASIS STUDIES

In 1992, technical basis studies were initiated to develop a design for the new operating requirements. The purpose of these studies were to 1) improve the ability to remove cadmium, chromium, and mercury contaminants from spent HEPA filters, 2) improve filter media retention, 3) consider waste minimization, and 4) improve filter processing rates. Twelve HEPA filters contaminated with cold calcine were used to evaluate and optimize 1) leach solution temperature, 2) nitric acid concentration, 3) multiple leach cycles, 4) acid recirculation, 5) filter orientation, and 6) air sparging. Understanding the effect of these variables allowed the system to be modified for optimum removal of cadmium, chromium, and mercury. As a result of the tests the HEPA Filter Leach System was modified to 1) reduce nitric acid concentrations from 4-8 molar to 1-3 molar, 2) utilize more leach cycles with shorter leach cycle times, 3) decrease the rinse cycle times, 4) orient the filters loaded-side down, 5) process HEPA filters one at a time, 6) utilize a new smaller process vessel to minimize leach solution volume, 7) utilize a new basket design for loose fiber retention, and 8) incorporate a new drying vessel that is separate from the process vessel (2).

VALIDATION TRIALS

The modified HEPA filter leaching concept was validated in 1994 by leaching six spent radioactive NWCF HEPA filters in the modified Filter Leach System. Representative filter media samples were collected and analyzed to show that the filters were no longer characteristically hazardous. The test results are shown in Table I (3). These analyses tested TCLP metals and selected volatile organic levels for waste codes that correspond to substances that are not "soluble to at least 5 Wt% in the water or an emulsion as applicable" (57 FR 37230, August 18, 1992). The results of these tests indicated that the volatile organic contamination was undetectable (<10-50 ppb). In addition to the reduction in hazardous constituents, the radioactivity dropped from 5-120 R/hr (β/γ) by a factor of 1000 to <100 mR/hr (β/γ) with a transuranic content of 0.4-3.6 nCi/g. Hence, the HEPA filter was no longer considered hazardous and could be disposed of as low-level radioactive waste.

Table I. Six HFLS filter media samples analyzed for RCRA metals in 1994.[a,b]

Constituent	Regulatory Limit (mg/1)	Filter #1 94-022110 (mg/1)	Filter #2 94-022512 (mg/1)	Filter #3 94-030418 (mg/1)	Filter #4 94-031125 (mg/1)	Filter #5 94-03169 (mg/1)	Filter #6 94-032822 (mg/l)
Arsenic	5	<0.170	<0.681	<0.671	<0.170	<0.170	<0.170
Barium	100	0.240	3.62	<0.865	<0.219	0.48	<0.219
Cadmium	1	<0.216	<0.866	<0.853	<0.216	<0.216	<0.216
Chromium	5	<0.208	<0.833	<0.820	<0.208	<0.208	<0.208
Lead	5	<0.143	<0.575	<0.566	<0.143	<0.143	<0.143
Mercury	0.2	<0.0402	<0.0402	0.018	0.016	0.092	0.012
Selenium	1	<0.149	<0.599	<0.590	<0.149	<0.149	<0.149
Silver	5	<0.194	<0.777	<0.765	<0.194	<0.194	<0.194

a This table provides the analytical results for each RCRA TC Metal analyzed for in the filter media samples. Detection limits will vary based on the size of the sample dilution.

b These analyzes were performed at INTEC laboratories.

Testing results were submitted to the Idaho Department of Environmental Quality and forwarded to the United States Environmental Protection Agency (USEPA) for a Determination of Equivalent Treatment (DET). In May of 1996 the USEPA determined that a DET is not necessary or applicable for the HEPA Filter Leach System (5). It was determined that treatment of HEPA filters in the HEPA Filter Leach System should be regulated as a hazardous debris as presented in 40 CFR 268.45 Table 1 "Alternative Treatment Standards for Hazardous Debris". Disposal of the spent solvents to the tank farm for subsequent calcination was determined to be constant with current operating practices. Since then the NWCF HEPA Filter Leach System has been used to process 78 filters in 1997 and 1998.

HEPA FILTER LEACH SYSTEM DESCRIPTION

The existing filter leaching system is shown in Figure 1 and consists of a leaching vessel, drying vessel, two work tables, associated piping, and instrumentation. The leach vessel, drying vessel, and work tables are located in the filter handling cell. Piping lines pass through the cell wall to utility service lines in the clean operating corridor. Remote handling capabilities are available within the cell via a pair of master-slave manipulators located on either side of the window, an overhead crane, and an electro-mechanical PaR[*] with a robotic arm.

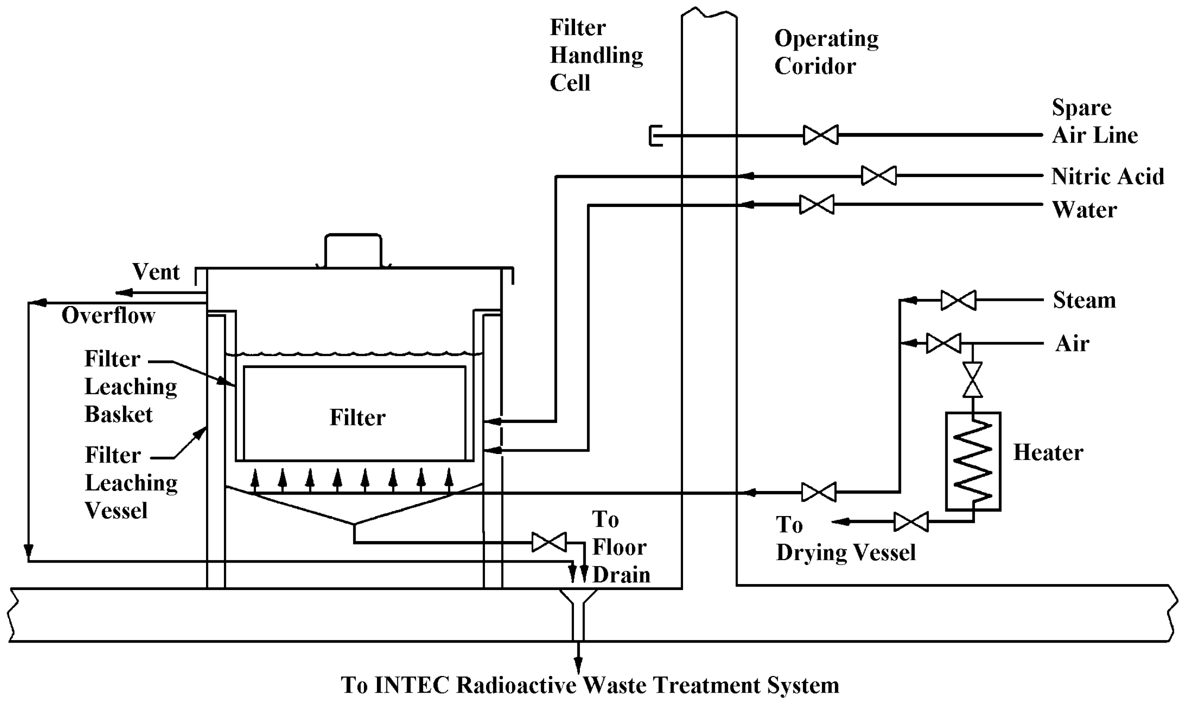

Figure 1 HEPA Filter Leaching System

The filter handling table is located behind the leaching vessel and is directly under the filter handling cell hatch cover. The table is constructed of stainless steel with an open grating working surface. It serves as the primary work surface for filter handling operations.

Contaminated HEPA filters are transferred into the filter handling cell through the PaR parking area and shielding doors using the filter handling tool and decon cell crane. A contaminated filter is placed in a basket which completely encompasses the filter to retain loose media fibers. This is to prevent vessel drain plugging during the leaching process. The top and bottom of the basket are constructed of 14 mesh stainless steel screens with 1.35 mm (0.053 inch) openings to retain the loose filter media. The open leach basket is placed on the filter handling table. The filter is placed inside the top half of the leaching basket so that the stainless screen side is facing upward. The lid of the basket is then closed and locked using the cell crane, PaR, or manipulator. The leach basket and filter are then placed on the basket support inside leaching vessel.

A mixing vessel is used to heat 1-2 M nitric acid to 43ºC (110ºF). The volume of the acid per leach cycle is approximately 227 L (60 gallons) which is enough to submerge the HEPA filter. The leaching operation begins when the acid solution is transferred to the leach vessel and heated by steam to 88ºC (191ºF) keeping the vessel lid closed. Air sparging at 10 scfm is initiated immediately. The filter is leached for 30 minutes. The leachate waste solution is then drained to a collection vessel. Second and third leach cycles follow the first leach cycle. After completion of the three leach cycles, two water rinses are conducted at ambient temperature for 10 minutes each. The volume of water per rinse cycle is approximately 227 L (60 gallons).

After completion of the leaching and rinsing cycles, the filter is allowed to drain for 10 minutes or until no more liquid drips out of the filter. Then the filter is transferred to the drying vessel. The drying operation is performed by forcing air through an air heater to raise the air temperature to 149ºC (300ºF). The air is then forced into the bottom of the vessel at 60 SCFM, through the filter, and out to the vent at the top of the drying vessel. Drying time is usually 4 to 6 hours.

After the filter is dried, the leaching basket containing the leached filter is removed from the drying vessel by the cell crane and placed on the filter handling table. The basket is rotated upside down so that the fiberglass screen is upward and the lid of the leach basket is then opened. The filters are sampled on a random basis to verify that the leaching process is performing adequately. The filters are then stored in the cell until enough filters have been accumulated to fill a wooden waste box for disposal.

ONGOING DEVELOPMENTAL ACTIVITIES

The Filter Leach System has been successful in reducing radionuclides and hazardous materials on the HEPA filters to allow them to be disposed of as low-level radioactive waste. However there are a few problems that are the focus of ongoing developmental activities. These activities include 1) investigate methods to prevent the HEPA filters from falling apart during the leaching process, 2) improve rinsing methods to minimize the amount of contaminated solution retained in the filter media, 3) improve fluid agitation systems to increase solubility rate and minimize damage to the filter media, 4) tools that would facilitate process manipulations or minimize cross contamination during process manipulations, 5) investigate methods that would reduce the amount of time required to process a HEPA filter thus improving process throughput, and 6) investigate solvents that will minimize or eliminate aqueous waste going to the Tank Farm.

CONCLUSION

The Idaho National Engineering and Environmental Laboratory HEPA Filter Leach System has successfully treated 78 HEPA filters classified as a TRU mixed waste in accordance with the "Alternative Treatment Standards for Hazardous Debris". Radioactivity was reduced from 5-120 R/hr (β/γ) to 30-90 mR/hr (β/γ) while TRU levels were dropped from over 600 nCi/g to 0.4-3.6 nCi/g. Cadmium, chromium, and mercury concentrations on spent HEPA filter media were successfully reduced to below disposal limits set by the Resource Conservation and Recovery Act (RCRA). The treated HEPA filters were disposed as low-level radioactive waste.

FOOTNOTES

* Mention of specific products and/or manufacturers herein implies neither endorsement or preference, nor disapproval by the U.S. Government, any of its agencies, or Lockheed Martin Idaho Technologies Company of the use of a specific product for any purpose.

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