

DEVELOPMENT OF A PULP PROCESS FOR TREATING CONTAMINATED HEPA FILTERS (III)

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ABSTRACT

The Pulp Process (PP) Treatment option was conceived as a replacement for the current Filter Leaching System (FLS). The FLS has operated at the Idaho Nuclear Technology and Engineering Center of the Idaho National Engineering and Environmental Laboratory since 1995 to treat radioactive, mixed waste HEPA filters. In recent years, the FLS has exhibited difficulty in removing mercury from the HEPA filters as the concentration of mercury in the spent HEPA filters has increased. The FLS leaches and washes the whole filter without any preparation or modification. The filter media and the trapped calcine particles are confined in a heavy filter housing that contributes to poor mixing zones around the edges of the filter, low media permeability, channeling of the liquid through cracks and tears in the filter media, and liquid retention between leach and rinse cycles. In the PP, the filter media and the trapped calcine particles are separated from the filter housing and treated as a pulp, taking advantage of improved contact with the leach solution that cannot be achieved when the media is still in the HEPA filter housing. In addition to removing the mercury more effectively, the PP generates less volume of liquid waste, requires a shorter leach cycle time, and possesses the versatility for treating filters of different sizes.

A series of tests have been performed in the laboratory to demonstrate the advantages of the PP concept. These tests compare the PP with the FLS under controlled conditions that simulate the current operating parameters. A prior study using blended feed, a mixture of shredded clean HEPA filter media and non-radioactive calcine particles, indicated that the PP would significantly increase the calcine dissolution percentages.

In this study, hazardous-metal contaminated HEPA filter media was studied. The results of side-by-side tests indicated that the PP increased the mercury removal percentage by 80% and might be a solution to the mercury removal problem encountered by the current FLS. A patent application has been filed for the PP and the patent is pending. In order to validate the PP and collect information for engineering design and economical feasibility studies, pilot plant scale tests are planned.

The PP requires the filter media to be removed from the filter housing. This can be accomplished by shredding the entire filter or by removing/shredding the filter media only. A "wire whip" concept was proposed to shred/separate the filter media from the housing by putting a wire whip in the center of the filter. The whip would be applied basically as a mixer. Discussions of the filter shredding and the wire whip concept are included in this paper.

INTRODUCTION

Background

The Idaho Nuclear Technology and Engineering Center (INTEC) at the Idaho National Engineering and Environmental Laboratory (INEEL) was operated from 1953 to 1991 to process spent nuclear fuel by dissolving the fuel and extracting the reusable uranium. Liquid waste from the fuel reprocessing was calcined at the New Waste Calcining Facility (NWCF) to form a dry powder, and put into concrete storage bins for storage. NWCF operations involved the handling of fission products, transuranic (TRU) materials, and hazardous materials. High Efficiency Particulate Air (HEPA) filters were used in the NWCF off-gas streams to prevent these materials from entering and contaminating the environment. Reprocessing and waste treatment have generated many HEPA filters contaminated with highly radioactive (5-120 R/hr (β/γ)), transuranic (TRU content >600 nCi/g have been observed), and hazardous metal constituents (6-12 mg Cd/g media, 2-4 mg Cr/g media, and 3-6 mg Hg/g media) (1-3).

Currently, there is no disposal facility that will accept the contaminated HEPA filters as generated. Hence, the Filter Leach System (FLS) was designed to lower radiation and contamination levels and reduce the cadmium, chromium, and mercury concentrations on spent HEPA filter media to meet disposal criteria set by the Resource Conservation and Recovery Act (RCRA). The treated HEPA filters are disposed of as low-level radioactive waste.

The original FLS concept was demonstrated in 1984, followed by the construction in 1988 (1). The technical basis for the system was established and optimized in initial studies using simulants in 1992 (2). The treatment concept was validated for the Environmental Protection Agency (EPA) approval in 1994 using six spent, NWCF HEPA filters. Post-leach filter media sampling results for all six filters showed that radioactivity was reduced from 5-120 R/hr (β/γ) to 30-90 mR/hr (β/γ) while TRU levels were lowered from over 600 nCi/g to 0.4-3.6 nCi/g (3). Cadmium, chromium, and mercury concentrations on treated HEPA filter media were successfully reduced below Land Disposal Restriction (LDR) limits set by RCRA (3).

The FLS was approved by the EPA and the State of Idaho for the treatment of HEPA filters at INTEC in 1996 (4). According to the approved procedure, leaching of each unit 2'x2'x1' filter requires three 60-gallon nitric acid solution (1-3N) leach cycles followed by two 60-gallon water rinse cycles. This generates 300 gallons of dilute nitric acid waste per filter. The FLS has processed 177 HEPA filters since 1997.

The FLS has performed very well in the past. However, as the concentration of mercury in the Calciner off-gas system has increased in recent years, more mercury has found its way to the HEPA filters. The FLS has had some difficulty removing mercury at these higher concentrations. In addition to performance issues, the HEPA FLS has the following shortcomings: 1) excessive volume of liquid waste generation that impacts INTEC waste minimization/elimination goals, 2) undesirable corrosive solution sent to Process Equipment Waste (PEW) tanks; 3) long leach cycle time, and 4) low versatility for treating filters of different sizes.

The Pulp Process

It was determined that the effectiveness of the existing FLS could be improved by finding a more efficient leach chemical to improve contaminant dissolution and/or developing a process to improve mass transfer rates. These two concepts led to the development of the Pulp Process (PP), for which a nonprovisional patent application has been filed and the patent is pending (5-8).

The PP improves mass transfer rates by removing the filter media from the restrictive filter housing and shredding the filter media to a finely divided pulp. It takes advantage of improved/additional physical contact that cannot be achieved when the media is still in the HEPA filter housing. The process eliminates the problem of channeling through cracks in the media and inefficiencies observed in the current FLS. The process improves circulation of the fluid through the contaminated filter media for improved contaminant removal while minimizing the amount of chemicals required. In addition, the pulp media can be de-watered more efficiently using vacuum/pressure filtration techniques thus minimizing the liquid drag-out problems observed with the existing system. The PP can handle multiple HEPA filters and filters of different sizes.

Scope

This study is to use hazardous-metal contaminated HEPA filter media as feed to further demonstrate the advantages of the PP over FLS through a series side-by-side test.

EXPERIMENTAL

Apparatus

Two sets of apparatuses were used in this study. A separate apparatus was used for the PP tests than for the FLS. The PP test apparatus consists of a 500-ml, round bottomed, 3-neck flask with a heating mantle. The central neck accommodates an overhead agitation shaft, the second neck holds an electronic thermometer and the third holds a condenser (5-6).

The "sandwich" tests, which simulate the FLS, were conducted using a specially designed reaction vessel. This vessel is a stainless steel cylinder with an inside diameter of 2³/₄" and a total height of 7". A 16-mesh screen is fixed inside the cylinder at a position 1¹/₂" from the bottom. This fixed screen and another 16-mesh movable screen placed on top of the samples were used to support and secure the samples during treatment. This configuration is very similar to what is currently used in the FLS facility, where the HEPA filter is placed in a leaching basket that has screens on the top and bottom to retain loose filter fibers. A 1/8" tube is connected to the reaction vessel 1/2" from its bottom to introduce sparging air during dissolution process. Both the bottom and the top of the reaction vessel are closed with plastic lids. On the top lid, there are two openings, the large one in the center to accept a condenser, and the small one on the side for a thermometer probe. The reaction vessel is put in a water bath with constant temperature control (5-6).

Filter Media

The contaminated filter media feed, which is the filter media of a HEPA filter removed from the off-gas duct of the 10cm Calcination Pilot Plant located at INTEC was used in this study. The filter is contaminated with hazardous metals, such as cadmium, chromium and mercury but have no radioactive or TRU constituents. The leaching efficiency of the process was measured by the metal removal percentage (MRP) as described in the following section.

Because the contaminant distributions along the width, length, and depth are not uniform, five sampling points were selected for the HEPA filter: one at the center (Point A) and one at each corner (Point B, C, D and E). A cylindrical filter media core was taken from each sampling point by cutting through the HEPA filter media with a special tool (9).

The 1-foot long cylindrical filter media core was then cut into four sections, labeled as Bottom (B), Mid-Bottom (M-B), Mid-Top (M-T), and Top (T) in the order from bottom to top respectively. From each core, one section was used for "sandwich" tests, one section was used for PP tests, and the other two sections were held as reserves.

The samples used for the PP and "sandwich" tests were two sections adjacent to each other. For example, if at one sampling point, the section B was taken for the PP test, then the section M-B was used as the "sandwich" test sample. Also the section for PP test and the section for "sandwich" test alternatively reversed the up-down position. That is, at points A, C, and E, the sections for PP tests were located above the sections for "sandwich" test; at point B and D, the sections for PP tests were located beneath the sections for "sandwich" tests.

PROCEDURE

Pulp Process Tests

Nine grams of shredded contaminated filter media was blended with 250 ml of 2N nitric acid solution at the temperature of 88 °C for 0.5 hour. The calculated solid to liquid ratio (S/L) was 0.036 g/ml, same as that used in the FLS.

When the allotted reaction time was complete, the filter media was separated from the leaching solution by filtration. The filter media residue was dried in an oven overnight at 60 °C and weighed. The volume of the filtrate was measured. Samples of filter media and filtrate residue were sent for analysis of mercury, cadmium and chromium.

"Sandwich" Tests

A core sample of the contaminated filter media sample weighing 14.4 grams was placed inside of the reaction vessel on the fixed screen. The movable screen was then placed above the sample. The reaction vessel was filled with 400-ml dissolution solution at 88 °C and the purging air was turned on with a flowrate of 460 ml/second for 0.5 hour. The calculated solid to liquid ratio (S/L) was 0.036 g/ml, same as that used in the FLS.

The filter media residue was collected, separated from the leaching solution by filtration and dried in an oven overnight at 60 °C. The filter media residue was weighed and

shredded and the volume of filtrate was measured. Samples of the shredded filter media residue and the filtrate were sent for analysis of mercury, cadmium and chromium.

CALCULATION

The metal removal percentage (MRP) was calculated with the following formula:

$$\text{MRP (Me)} = 100 \times \frac{V (\text{leachate}) * \text{Me (leachate)}}{V (\text{leachate}) * \text{Me (leachate)} + \text{Me (residue)} * W (\text{residue})}, \%$$

Where,

Me - Hazardous metal, Hg, Cd, or Cr

V (leachate) - Volume of the leachate, liter

W (residue) - Weight of the filter residue, Kg

Me (leachate) - Assay of the hazardous metal (Hg, Cd, or Cr) in the leachate, mg/liter

Me (residue) - Assay of hazardous metal (Hg, Cd, or Cr) in the filter residue, mg/Kg

RESULTS AND DISCUSSIONS

The average MRP and the MRP at each of the five sampling points for mercury, cadmium, and chromium are given in Figure 1, Figure 2 and Figure 3, respectively. At a reaction time of 0.5 hours, the average mercury MRP for the "sandwich" process is 40.9 % versus 74.0% for the PP, an improvement of 80.9%. This result indicates that PP could be a suitable alternate process to the mercury removal problem encountered by the current FLS.

The PP outperformed the "sandwich" process for cadmium removal by 24.1% and for chromium removal by 7.6%. The improvements for the MRP of these two metals are not as significant as that of mercury. Since the current FLS has no problem removing cadmium and chromium thus far, it is only an added benefit.

The laboratory experiments performed to date demonstrate that the PP could significantly improve the HEPA filter leaching efficiency, but did not fully explore the other advantages of the PP. First, the "sandwich" apparatus possesses some features more favorable than that of the FLS in terms of mass transfer and agitation. Referring to Figure 4, the filter paper inside the HEPA filter is continuously folded vertically as shown in Part I, so there is no opening in the filter media either at the top or the bottom end for the sparging air and leaching/washing liquid to flow through.

Samples for the "sandwich" test were taken from each of the four sections of the cylindrical filter media core, as shown in Part II of Figure 4. For section T, there are openings at its bottom end. For section B, there are openings at its top end. For section M-T and M-B, there are openings at both ends. Liquid flow and air dispersion inside the "sandwich" apparatus was probably much better than that experienced by the FLS, resulting in higher leaching efficiencies.

Second, one of the advantages of the PP is that the pulp media can be de-watered more efficiently using vacuum/pressure filtration techniques. During our laboratory PP experiments, the shredded filter media was separated from the leachate by gravity

filtration. The filter cake retained a significant amount of leachate after filtration, resulting in lower leaching efficiencies than what would be expected in an actual process.

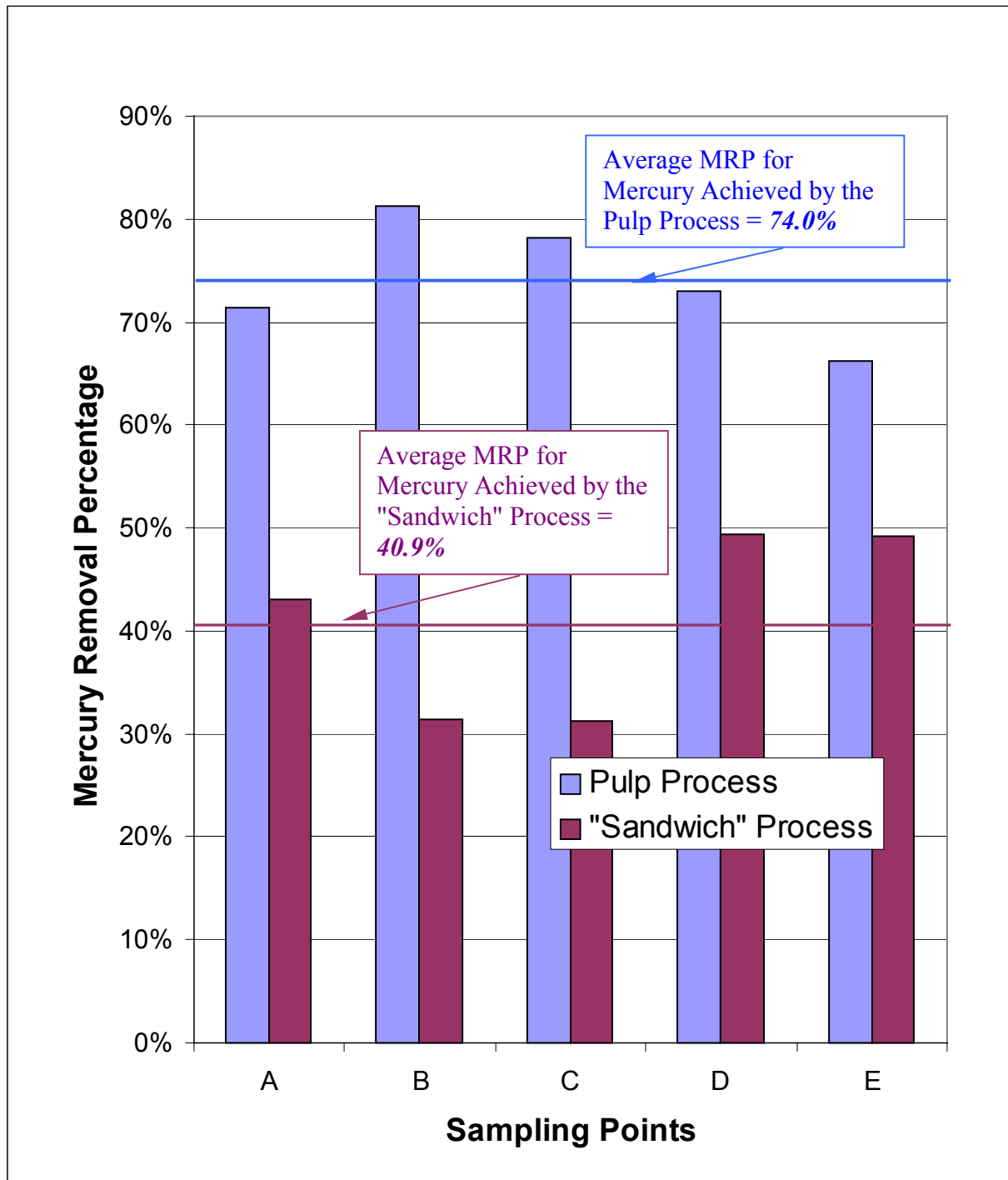


Fig. 1. Comparison of the Mercury Removal Percentages Achieved by the Pulp Process and the "Sandwich" Process

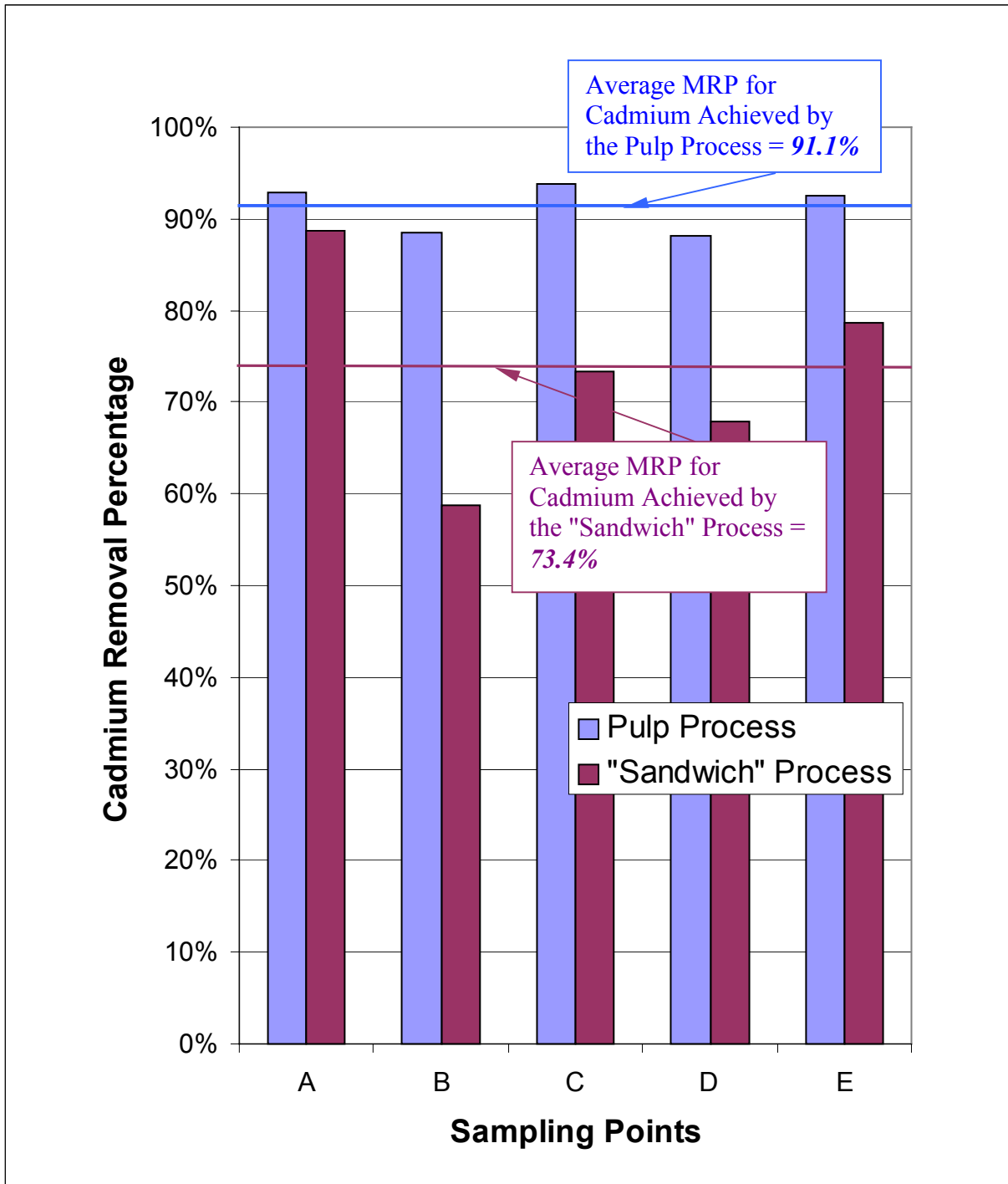


Fig. 2. Comparison of the Cadmium Removal Percentages Achieved by the Pulp Process and the "Sandwich" Process

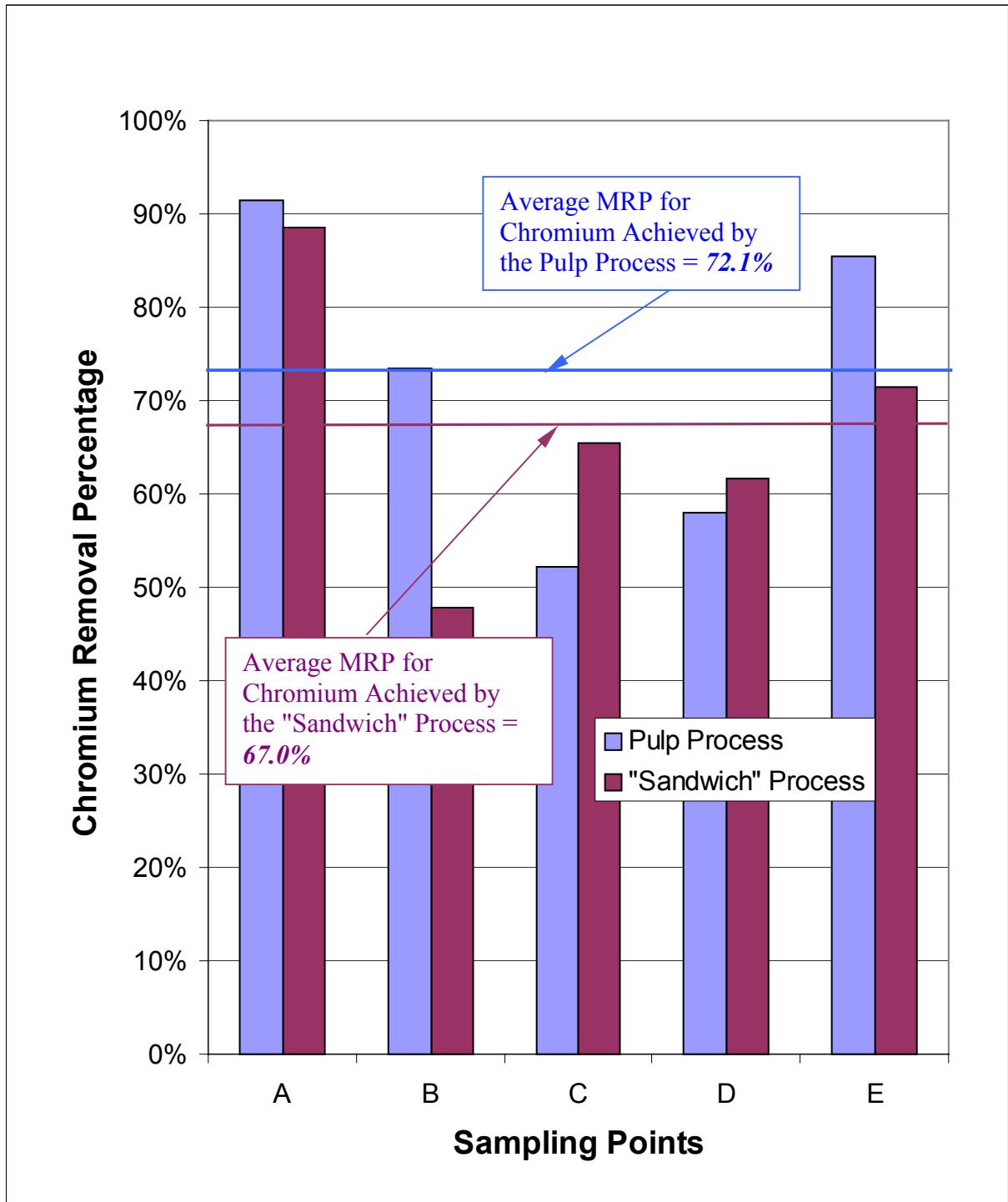


Fig. 3. Comparison of the Chromium Removal Percentages Achieved by the Pulp Process and the "Sandwich" Process

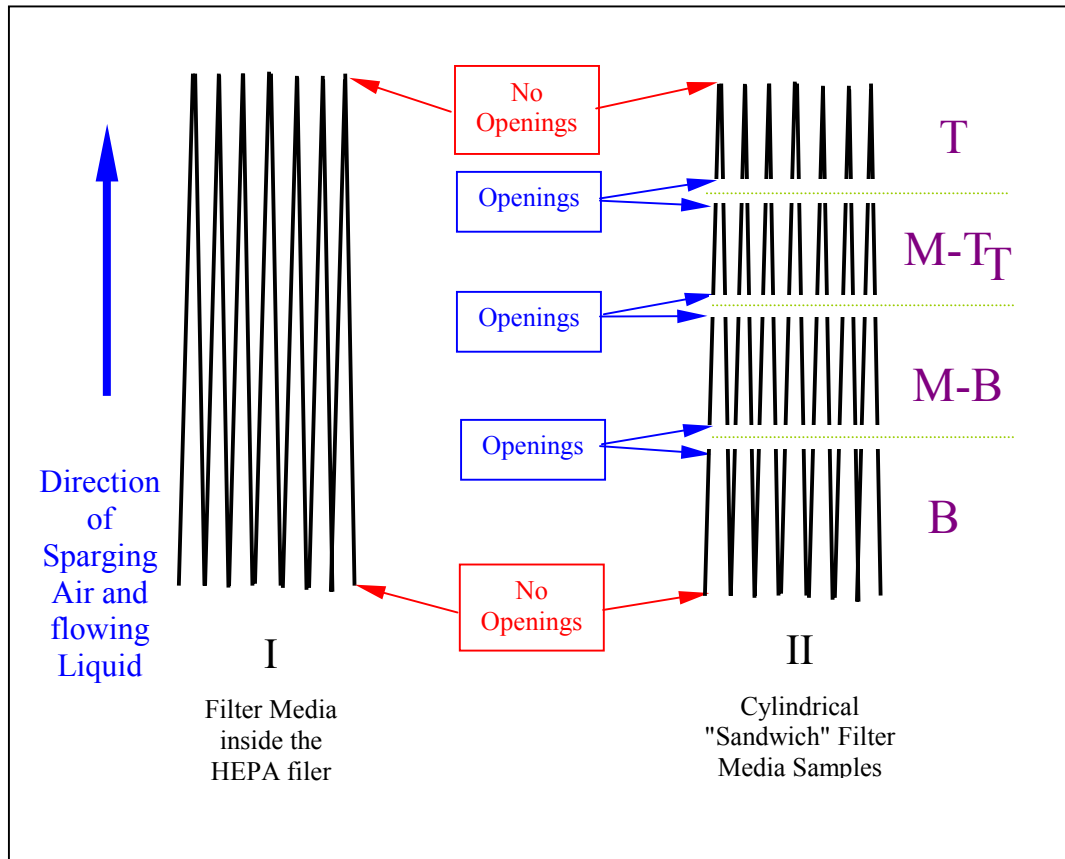


Fig. 4. Schematic Representation of the Filter Media Sheet Configuration inside a HEPA Filter and of the "Sandwich" Samples

Hence, the improved air/liquid flow rates for the laboratory "sandwich" tests and the lack of de-watering for laboratory PP tests, likely minimized the advantages of the PP. To avoid the above two pitfalls, pilot plant tests using the whole depth of the filter media and equipped with vacuum/pressure filtration need to be conducted.

HEPA FILTER SHREDDING

The separation of the filter media from the filter housing for the PP can be accomplished by either shredding the entire filter or by shredding/removing the filter media only.

Entire Filter Shredding

The available shredding technologies were evaluated with focus on products of the SSI Shredding Systems, Inc. A demonstration of SSI style four-shaft shredder using the steel-framed HEPA filters sent from the INEEL was conducted. The system shredded the filter into pieces of approximately 1/2 " by 1/2 ". According to SSI, if the electric motor is replaced by a double hydraulic motor, the shredded pieces would be smaller, the

shredding would be faster, and the dimensions of the equipment would be significantly reduced as well. Further size reduction is necessary before this equipment could be deployed for the remote handled filters at INTEC because this equipment is required to fit within the current decon cell at INTEC.

Filter Media Removing

A high-pressure spray nozzle can be used to cut the filter media from the filter housing. Preliminary work on filter media removal was evaluated (2, 10). Unfortunately, the filter media is well attached to the filter housing and is not easily removed. It was concluded that the nozzle was not favorable for remote operation and the investigation was discontinued.

A press could remove most of the filter media; however, some residual media could stay attached to the wall. Although a wire knife could be used to cut the remaining residual from the filter housing, this operation would probably be difficult to do remotely.

One of the options is a flexible, articulated bladed impeller. This alternative is to place a cutting fan in the vessel. A 2 ft impeller could be expected to remove about 3.14 ft³ volume from the center of the 2×2×1 ft filter (4 ft³). Therefore, 21.5% of the filter would remain attached to the filter housing.

The wire whip is another option conceived at the INEEL. The wire whip consists of a stainless steel wire in the center of a metal tube. A spinning air motor could be mounted on the lid of the current dissolution vessel. A guide on the lid would direct the whip to the center of the HEPA filter. The tube would be bent so that the wire extends out of the tube perpendicular to the length of the metal tube. When the tube is rotated and the wire starts to extend, more and more of the filter media would be shredded. The air motor could be located on a drill press device. The range of the press is from the bottom of the filter to the top of the filter. The position of the air motor could be controlled by a timer to go up or down at a continuous rate or pattern. The whip wire should be capable of removing 95% of the filter media from the filter housing regardless of the shape of the filter.

As the filter media is shredded, it either falls to the bottom of the shred vessel or is sucked by a vacuum to a media dissolution vessel for acid addition. If the operation of the shredder causes a dust problem, the shred system could be located in a sealed container. During shredding, leaching solution could be sprayed onto the filter media.

CONCLUSIONS

The results of laboratory experiments indicated that the Pulp Process is 80.9% more effective in mercury removal than a simulated FLS (“sandwich” process). In addition, the PP outperforms the simulated FLS in cadmium removal by 24.1% and in chromium removal by 7.6%. Thus, the PP is a potential replacement for the current FLS.

This increase in removal percentage should resolve the current problems with high mercury concentrations in the current FLS. The current system fails mercury limits at a toxicity characteristic leaching procedure (TCLP) for mercury (0.2 mg/g). In the past five years, since the initial studies on the FLS, more mercury has been seen in the

processing streams and in the off-gas systems at the NWCF. The initially reported decontamination factors for the FLS was about 65 (1). This means that for every milligram of mercury that goes into the system, approximately 15 micrograms of mercury remains. If the decontamination factor were increased by 80% to 117, this would allow the current fundamental limit of 13 mg/g of mercury in the unleached filter to be increased to 23.4 mg/g. This would allow the over-limit filters to pass, as they are currently estimated to be about 13 mg/g mercury.

The laboratory experiments performed to date demonstrate that the Pulp Process will significantly improve the HEPA filter leach efficiency. Pilot plant tests are recommended to fully explore the advantages of PP over FLS and to collect information for the engineering design and an economic feasibility study.

Shredding the entire filter shredding to separate the filter media from the filter housing is not recommended. Among the alternative filter media removal options, the wire whip is believed to be the most promising one. More R&D work is needed to verify its applicability.

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