

**Development of Ultrafiltration and Inorganic Adsorbents for
Reducing Volumes of Low-Level and Intermediate-Level
Liquid Waste**

Quarterly Report

October, November, December, 1976

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MONSANTO RESEARCH CORPORATION

A Subsidiary of Monsanto Company

MOUND LABORATORY

Miamisburg, Ohio


45342

operated for

**UNITED STATES ENERGY RESEARCH
AND DEVELOPMENT ADMINISTRATION**

U. S. Government Contract No. EY-76-C-04-0053

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED



DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

DISTRIBUTION

External

J. A. Chacon, ERDA/DAO (2)
D. Davis, ERDA/ALO (2)
R. A. Wolfe, ERDA/DWMPR (2)
T. K. Keenan, LASL
J. O. Bloemke, ORNL
A. K. Williams, AGNS
R. E. Lerch, HEDL

Internal

A. G. Barnett, A-223
W. H. Bond, SM
W. T. Cave, OS-102
J. W. Doty, SM
K. V. Gilbert, OS-120
L. V. Jones, OS-128
D. P. Kelly, OS-106
J. W. Koenst, R-5
B. R. Kokenge, SM
D. F. Luthy, SM
D. L. Prosser, R-63
L. B. Stevens, OS-128
R. E. Vallée, OS-107
H. L. Williams, A-151
Library (15)
Technical Publications (3) for
 R. K. Flitcraft, MRC-Dayton
 J. R. McClain, A-231
File

DEVELOPMENT OF
ULTRAFILTRATION AND INORGANIC ADSORBENTS FOR
REDUCING VOLUMES OF LOW-LEVEL AND INTERMEDIATE-LEVEL
LIQUID WASTE

QUARTERLY REPORT
OCTOBER, NOVEMBER, DECEMBER 1976

INTRODUCTION

The objective of this program is to develop and demonstrate separation methods for removing radionuclides from liquid process waste streams. As part of this program, Mound Laboratory will develop lower cost alternatives for use in 1980 fuel reprocessing and waste solidification plants, evaluate the processes within the nuclear fuel cycle which contribute to low-level and intermediate-level waste, and determine the feasibility of ultrafiltration, reverse osmosis, inorganic adsorbents and other separation concepts as additions to process design to reduce the generation of this type of waste. In the initial phase of this program, membrane equipment will be obtained from a commercial membrane manufacturer. After the pilot plant is installed, it will be checked out on cold feed in order to obtain initial flux and rejection data for comparison to data obtained later on in the program. After completion of the cold tests, the membrane pilot plant will be run on a combined contaminated feed emanating from showers, laboratory drains, janitorial sinks and decontamination in processing areas, as well as a laundry waste stream containing alpha-contaminated wastes. This combined waste stream contains only alpha contamination (uranium and plutonium). However, as part of this program, gamma activity will be added to the waste stream. These wastes will be representative of those streams found at fuel reprocessing plants, as well as various ERDA processing facilities such as Mound, LASL, Hanford, and Rocky Flats.

For the second part of the program, laboratory tests will be run on various adsorbents to evaluate their capacities for removing radionuclides. As part of this program, a technique for screening adsorbents developed at Mound Laboratory will be utilized.

Concurrently, a system to conduct engineering tests will be designed and built. Engineering tests will then be conducted on several promising adsorbents chosen from the laboratory tests. The engineering tests will be run in columns in

order to determine various design parameters (i.e., loading ranges, bed capacity, throughput rates, residence times, velocity, and bed height). At the conclusion of this part of the program, data from several promising adsorbents will be utilized in order to design a pilot plant. Mound Laboratory will work closely with industry and ERDA facilities in the preparation of typical waste compositions and ultimate process design.

ULTRAFILTRATION

The work done on the ultrafiltration project since its inception on October 1, 1976, has been of a preparatory nature. Necessary equipment for a pilot plant has been ordered from ABCOR, Inc., and is scheduled to be installed by April 20, 1977. Work on evaluating the effect of radioactivity on membranes has been subcontracted to Walden Research. A literature search for pertinent information on ultrafiltration, reverse osmosis, and other related subjects is presently underway.

Work this quarter has been directed toward performing bench-scale tests with small ultrafiltration modules. Many runs have been made using the influent to Mound's waste disposal (WD) facility which consists of radioactive laundry wastes, and Plutonium Processing (PP) Building low-level solutions.

A series of laboratory experiments were run on WD influent using Romicon hollow fiber ultrafiltration modules. These modules had various molecular weight cut-offs ranging from 2,000-80,000 MW. The rejection of conductivity was found to be low in most cases. The rejection of radioactivity ranged from 90-98%, depending on the membrane type and on the feed concentration. Typical product activity ranged from 7-100 dis/min/ml of alpha activity. Since feed concentrations changed daily, results tend not to be consistent. Also, a slight problem of leakage across the headers of the module, in some cases, tended to make the results inconsistent.

Experiments were also run on contaminated laundry waste streams. Results ranged from 98->99.8% rejection of activity, depending on the membrane type. This yielded a product concentration of <0.1 dis/min/ml of alpha activity.

Tests on contaminated floor drain wastes from the Plutonium Processing area yielded rejections of 85-88% alpha activity depending on the membrane type.

These experiments have shown that the ability to remove radioactivity is a function of the waste stream. This is caused by the fact that the activity in the waste water is in various forms (ionic, polymeric, colloidal and adsorbed into suspended solids). Removal of suspended or colloidal material was very high, while removal of ionic material was not as effective. Radioactive laundry waste proved to be the easiest to decontaminate, while the PP Building low-level solution was the most difficult. Decontamination of WD influent solution, which is a combined waste stream, varied considerably from day to day because of its constantly changing make-up.

Runs were also made using WD influent with various additions, such as polyelectrolytes, complexing agents, and coagulants. The purpose of these runs was to determine if these additives would aid in the removal of radioactive material from the various waste waters by complexing the ionic species. At the present time, none of the additives evaluated has had much effect, but experiments are continuing.

As stated above, some solutions were decontaminated better than others; however, in all cases, a rejection of activity of greater than 90% was obtained, and in some cases, a rejection of greater than 99% was achieved. Also, in some cases, rejection of suspended solids and colloidal material was nearly complete and the product solution was completely clear.

During this quarter, a technical report was being written, giving results of all the experiments discussed above. This report will be submitted as an MLM (Mound Laboratory Manuscript).

In order to identify the waste stream further, analyses were run to determine the levels of suspended solids, dissolved solids, and alpha activity of influent water at WD. The analysis showed that levels of suspended solids ranged from 2 or 3 mg/l to over 200 mg/l, and alpha radiation levels from 50 dis/min/ml to over 100,000 dis/min/ml, while dissolved solid levels ranged from 1,000 to 1,500 mg/l.

ADSORBENTS

Mound Laboratory is evaluating macroporous ion exchange resins for the removal of plutonium, uranium, and various colloids from process waste treatment effluents. A number of organic ion exchange resins were evaluated for $^{238}\text{Pu}(\text{IV})$, $^{238}\text{Pu}(\text{VI})$, and $^{233}\text{U}(\text{VI})$ removal from water using batch isotherm tests. The

capacity and equilibrium distribution coefficients were compared with each other and to bone char, an inorganic adsorbent consisting of hydroxylapatite (HAP). The various types of adsorbents showed that the extent of removal and equilibrium coefficients (Kd) were a function of pH. For removal of polymeric plutonium $^{238}\text{Pu}(\text{IV})$, the best results were achieved using the inorganic adsorbent, bone char (hydroxylapatite), at pH 7. This had been shown previously in work done at Mound Laboratory for the Division of Physical Research (ERDA). However, Amberlite XE270, a macroporous weak base anion exchange resin, also showed reasonable Kd values at pH 7. Therefore, the best removal of polymeric plutonium can be achieved using chemisorption or weak base anionic exchange (indicating strongly ionized anions). XAD2, a resin which removes solute by an adsorption process (hydrophobic), also showed some promise. That is, it performed better than the remaining resins, but not as well as bone char and XE270.

For removal of ionic $^{233}\text{U}(\text{VI})$, the strongly acidic cation exchangers gave the better results; the Kd values were on the order of 10^2 better than bone char. Again, performance was a strong function of pH. In most cases, pH 4 and 7 gave the better results. However, for anionic resins, the better results occurred at pH 7 and 10. Adsorbent resins which remove constituents by physical adsorption (XAD2 and XAD7) did not perform well for uranium removal. In comparing the strong base anion exchange resins, the gel resin, Dowex SBR-P, had much lower Kd values than the macroporous resin, Dowex MSA-1, thus showing the main advantage of using the new macroporous resins. In fact, for the Dowex resins, both cationic and anionic macroporous resins gave Kd values on the order of 10^5 at various pH's, while the gel resins gave Kd values of the order of 10^2 .

Excellent results for removal of $^{238}\text{Pu}(\text{VI})$, were achieved using a strong base macroporous anion exchange resin, Amberlite XE279. The Dowex-SBR-P strong base anion exchange gel resin gave Kd values lower than bone char, while XE279 gave Kd's a factor of 10 better than bone char. The best results were achieved using Amberlyst XN1010, which gave Kd's a factor of 20 better than bone char. The XN1010 is a strong acid cation exchange macroporous resin used mainly for catalysis in organic liquids, and also for cation removal from nonaqueous solutions.

During this period, a report was written detailing the work described above. The report (MLM-2320) was entitled, "Evaluation of New Macroporous Resins for the Removal of Uranium and Plutonium from Waste Streams", and was published December 20, 1976.

Small laboratory columns are being run in order to obtain data to design a system for engineering tests. The three (3) resins which showed the highest Kd values at each pH (4, 7, 10) during the batch tests were being run with solutions containing U, and Pu (IV Polymer & VI). The test solutions contain approximately 13,000 dis/min/ml and are being fed at a rate of 2 ml/min through columns of 0.7 cm I.D., with a resin height of 6-7 cm. Each column contains one gram of resin. Data from these tests will determine the resins which have the best breakthrough ratio to given weight (1 gm) for a dynamic system.

Work on the design and construction of the engineering test facility is continuing. The facility will be available for use, starting in May, 1977. The columns contained in this facility will be 2 inches I.D. and will contain a bed height of approximately 30 inches. Feed flow rates will be proportional to the rates determined using the small 0.7 cm I.D. columns, described previously.