LOW-LEVEL WASTE DATA BASE DEVELOPMENT--EPICOR-II RESIN/LINER INVESTIGATION--A PROGRAM REVIEW[a]

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ABSTRACT

This paper presents an overview of the titled program and gives the status of the work on resin degradation, resin solidification, and field testing of solidified samples. A brief discussion of some recent results also is included. Resin materials from EPICOR-II prefilters used during cleanup of Three Mile Island Nuclear Power Station are being examined to (a) develop a data base for low-level waste and (b) obtain information on survivability of waste forms composed of ion exchange media loaded with radionuclides and solidified in matrices of cement and Dow polymer. An unusual aspect of the investigation is the use of commercial grade resins which have been loaded with over five times the radioactivity normally seen in a commercial application. That dramatically increases the total radiation dose to the resins.

INTRODUCTION

The March 1979 accident at Unit 2 of the Three Mile Island Nuclear Power Station released approximately 2,120,000 L (560,000 gal.) of contaminated water to the Auxiliary and Fuel Handling Buildings. The water was decontaminated using a three-stage demineralization system called EPICOR-II, which contained organic and inorganic ion exchange media. The first stage of the system was designated the prefilter, and the second and third stages were called demineralizers. Fifty EPICOR-II prefilters with high concentrations of radionuclides were transported to the Idaho National Engineering Laboratory (INEL) for examination and storage before final disposal at the commercial disposal facility in the State of Washington. Examinations are being conducted by EG&G Idaho, Inc. on materials from several of those prefilters. A schematic of an EPICOR-II prefilter is shown in Fig. 1.

The U.S. Department of Energy (DOE) initiated examination of several prefilters under the EPICOR-II Research and Disposition Program in FY-1982. In FY-1984, the U.S. Nuclear Regulatory Commission (NRC) included this work as part of the Low-Level Waste Data Base Development--EPICOR-II Resin/Liner Investigation Program. That change resulted in more emphasis placed on investigating the requirements of 10 CFR 61 "Licensing Requirements for Land Disposal of Radioactive Waste"[1] by obtaining data on performance of waste in a disposal environment. Studies to obtain that data are based on the requirements of the NRC Office of Materials Safety and Safeguards "Branch Technical Position (TP) on Waste Form."[2]

This paper reviews those examinations, briefly summarizing progress/results to date and presenting future plans. A schedule (Fig. 2) is included which identifies significant events in the program.

PROGRESS/RESULTS TO DATE

The investigation is divided into four tasks, each addressing a different subject on waste disposal.[3] The tasks are resin degradation, resin solidification, field testing, and liner integrity. Materials from six EPICOR-II prefilters have been studied in the program. The first two samplings of resins from prefilters PF-8 and -20 have been analyzed for degradation due to irradiation, with results presented in several documents.[4-6] Studies of resin solidification using resins from prefilters PF-7 and -24 are progressing, with several interim reports available.[7-11] Field testing of solidified waste forms containing resins from PF-7 and -24 has been initiated recently using lysimeters located at Argonne National Laboratory-East (ANL-E) and Oak Ridge National Laboratory (ORNL). Examination of liners from prefilters PF-3 and -16 was completed in FY-1983 and reported in FY-1984 and -1985.[12,13] Annual reports also have been published describing results of the examinations.[14,15]

The following subsections briefly summarize the significant progress/results of the program to date.

Task 1--Resin Degradation

This task was initiated in mid-FY-1982, as shown in Fig. 2. Design requirements of equipment were formulated for removing resin samples from several EPICOR-II prefilter liners. The experimental requirements to conduct the task were defined in FY-1983, and all required capital equipment was designed and fabricated. The first resin samples were removed from two liners, PF-8 and -20, that year. The second resin samples were obtained from those two liners in early FY-1986.

Chemical and physical analyses of the first resin samples were initiated in FY-1984 and completed in FY-1985. A NUREG report (Ref. 4) describing the first analysis also was completed and released. Chemical and physical analyses of the second resin samples were performed in FY-1986. The NUREG report describing the second analysis is in preparation. Results of both analyses are discussed in a companion paper (Ref. 6) presented at Waste Management '86.

The study of degradation of EPICOR-II organic ion exchange resins at INEL correlates with the findings of other researchers,[16-21] and degradation has been identified in the EPICOR-II resins at a lower than predicted total integrated radiation dose (Ref. 18). The internal dose received by the organic ion exchange resins in EPICOR-II prefilters PF-8 and -20 was sufficient to initiate degradation. The degradation at the time of analysis was measurable, as can be seen

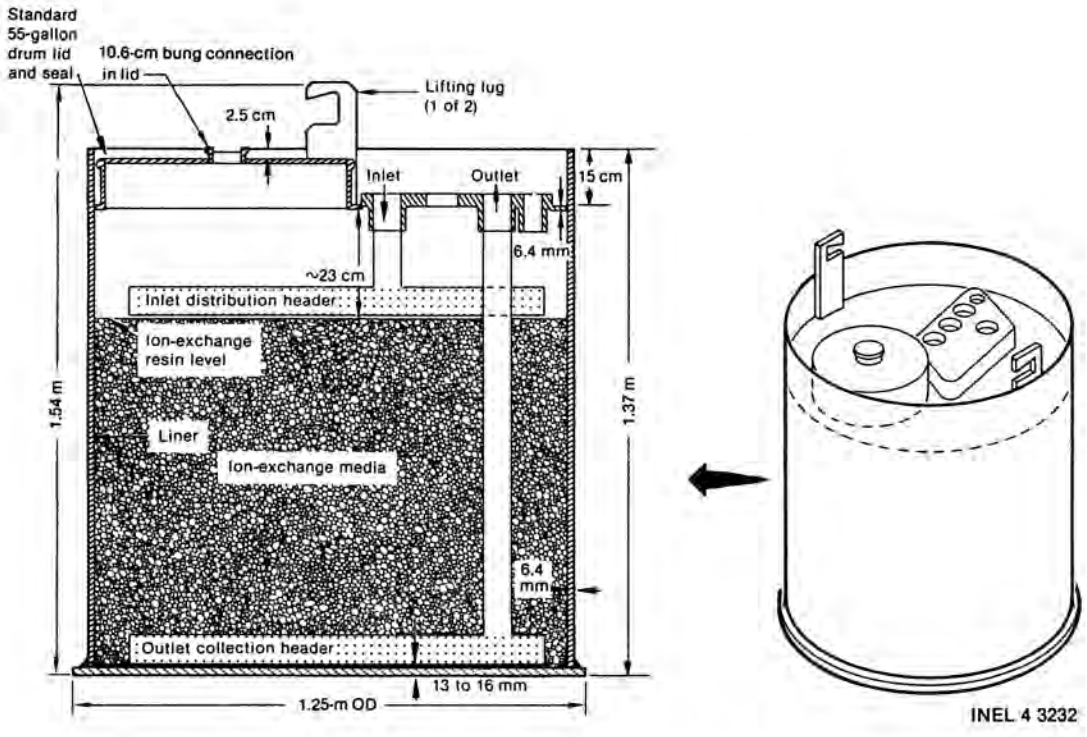


Fig. 1. Isometric and Full Section Schematic of an EPICOR-II Prefilter.

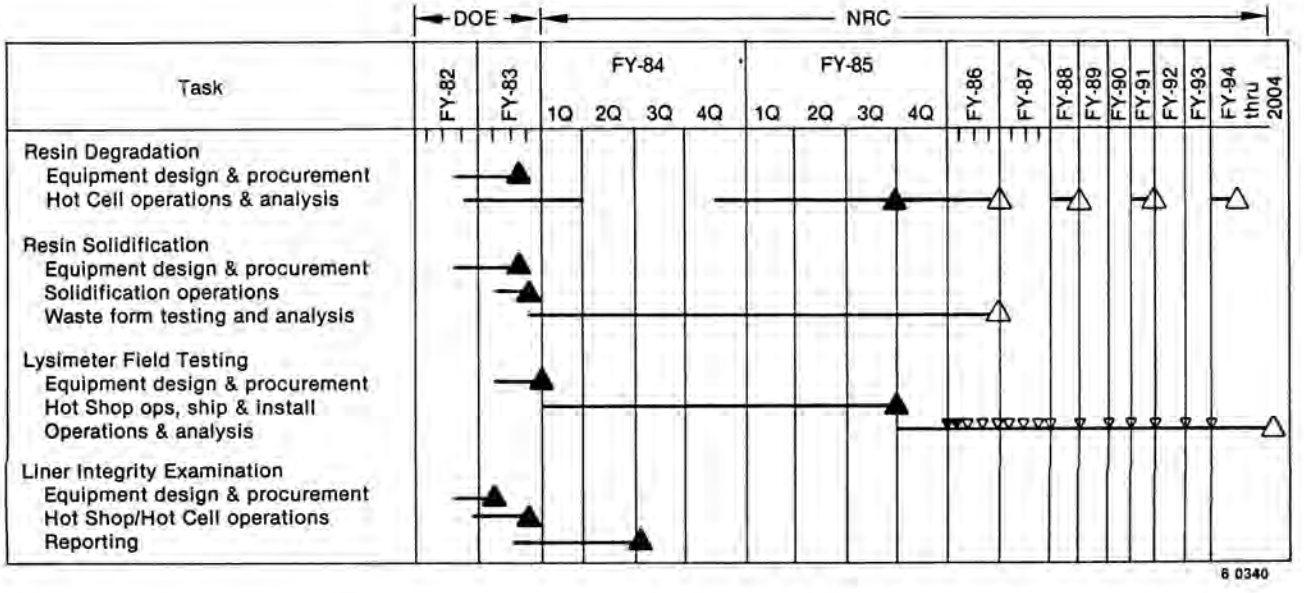
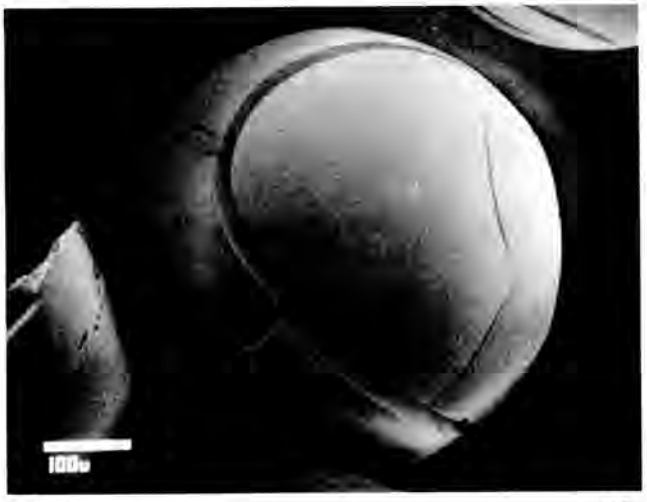


Fig. 2. Schedule of Significant Events in the Low-Level Waste Data Base Development--EPICOR-II Resin/Liner Investigation Program.

by the damage to the resins (Figs. 3 and 4). Also, the equilibrium of the polymer structure had been shifted toward polymer breakdown. Further sampling and analysis of EPICOR-II resins will help determine the rate at which the resins are being degraded.



PF-8#1 150x
6 0342

Fig. 3. Scanning Electron Microscope Photomicrograph Showing Damage (Curved Fractures) to the Strong Acid Cation Resin From PF-8 Due to Degradation.

Findings from the degradation study can be related to commercial disposal of spent ion exchange media used in power reactors. The determination of onset of degradation and, later, of significant loss

PF-8#2—Phenolic cation 80x
6 0339

Fig. 4. Scanning electron microscope photomicrograph showing damage (spongy appearance) to the phenolic cation resin from PF-8 due to degradation.

of exchange capacity (with resultant loss of radionuclides) will provide a data base useful in planning for, and regulation of, disposal of ion exchange resins.

Task 2--Resin Solidification

This task was initiated in mid-FY-1982, as shown in Fig. 2. Equipment used for removing resin samples was designed and fabricated under Task 1. The experimental requirements were identified in FY-1983; design requirements for the solidification equipment were completed; and all capital equipment was designed and fabricated.

Removal of the required resins from the two prefilters, PF-7 and -24, was completed and the resins were solidified into waste forms using either Portland cement or Dow polymer in FY-1983. Examination of waste forms was initiated in FY-1984. The solidified waste forms, each 4.8 cm (2 in.) in diameter by 7.6 cm (3 in.) long, are being examined in accordance with the TP to measure (a) compressive strength (initially, and after thermal stability, leachability, radiation stability, and biodegradability testing); (b) free standing liquids; and (c) homogeneity. A letter report was issued describing results of the initial compressive strength, free liquid determination, and homogeneity testing (Ref. 8).

Examination of waste forms for thermal stability, leachability, radiation stability, and biodegradability was conducted during FY-1985 and -1986. Results of the thermal stability and leachability tests were issued by letter reports (Refs. 9 and 10). A letter report on radiation stability testing is being written. Testing for biodegradability will continue through FY-1986. A more detailed account of the above work is presented in another companion paper (Ref. 11).

The objective of the resin solidification task is to confirm the adequacy of the test procedures recommended in the TP for ion exchange resins having high radionuclide loadings. Testing actual ion exchange media wastes provides an opportunity to assess how well the test methods in the TP ensure solidification into a stable product. If highly loaded ion exchange resins require alternative testing protocols, those methods will be recommended.

Portland Type I-II cement and Dow polymer waste form specimens (Fig. 5) incorporating EPICOR-II ion exchange resin wastes from PF-7 and -24 were found to meet the free standing liquid and compressive strength requirements of the TP. They also were resistant to thermal degradation, leaching, and immersion, as specified by the TP. The test results indicate the general applicability of ASTM B 553-79 and ASTM C 39-72 for thermal stability testing of waste forms, and ANS 16.1 and ASTM C39-72 for leach and immersion testing of waste forms, as specified in the TP. However, minor modifications to the TP are recommended to ensure that (a) evaporative water loss from specimens does not occur during or before thermal cycling, (b) times at temperature are sufficient to achieve desired maximum and minimum temperatures, (c) ANS 16.1 retest criteria are followed, and (d) the immersion fluid types are specified.

Task 3--Field Testing

This task was begun in mid-FY-1983. Preliminary experimental requirements were completed for lysimeter systems and waste forms. National laboratories were contacted to participate in the task. The field testing experiment was designed, and equipment required to conduct the task was placed on order by the end of FY-1983. Waste forms were molded using resins removed from PF-7 and -24 and mixed with either Portland cement or Dow polymer.

Fig. 5. Example of an EPICOR-II Resin Waste Form Solidified With Dow Polymer.

Two participating laboratories were selected, ANL-E and ORNL. Lysimeter systems and casks containing the waste forms were transported to the participating laboratories in early FY-1985. There are 10 lysimeters, 5 at ANL-E and 5 at ORNL. Four lysimeters at ANL-E were filled with local soil, and four at ORNL with soil having the characteristics of that from the waste disposal site at Barnwell, SC. One lysimeter at each site was filled with inert media. Procurement of all capital equipment was completed during FY-1985.

The lysimeter systems (Fig. 6) were installed in the laboratory waste areas; the waste forms, moisture cups, and detectors installed in each lysimeter; and the lysimeters filled with the specified soils or inert material. Seven waste forms were stacked end-to-end in each lysimeter to provide a 1-L volume.

Monitoring of moisture cups, designed to track migration of radionuclides, began with collection of liquid samples in September 1985 (three months from the time of placement) and will continue once every three months thereafter. The monitoring includes sampling of liquids from locations near the waste form and chemical analysis of those liquids. Soil moisture/temperature at three elevations in each lysimeter, along with a complete weather history, are recorded on a continuing basis by a data acquisition system. It is anticipated that radionuclide mitigation from the waste forms will not be observed for several years. Those findings will be issued in a letter report.

The objective of Task 3 is to compare results of the short-term laboratory leach tests in Task 2 with actual leaching in the field. If short-term leach tests do not compare well with the field tests, EG&G Idaho will recommend alternate leach testing methods.

Task 4--Liner Integrity Examination

Two prefilter liners were examined metallurgically in FY-1983. First, the ion exchange media from prefilters PF-3 and -16 were transferred into replacement liners. After visual examination of the liners, removal of metallurgical samples, and metallurgical evaluation of the removed samples, the liners were disposed. The GEND[a] report describing the results of the examination (Ref. 12) was released and distributed under DOE funding. The equipment design requirements were developed for this task, and most of the hardware was fabricated during FY-1982. All capital equipment was completed by early FY-1983. The task was terminated after distribution of the GEND report.

Visual examination of the liners revealed that the exterior and interior base metal surfaces appeared sound (Refs. 12 and 13). It also was noted that the interior coatings were blistered, loose, and in some locations had spalled or chipped. There appeared to be more coating failures in liner PF-16 than -3. The interior surface of liner PF-3 was coated with a thin, rust-colored film.

Corrosion products were found between the coatings and the base metal on both the interior and exterior surfaces of both liners. In general, the corrosion products and coatings adhered to the base metal. Areas were observed where corrosion products were surrounded by coating, which suggests that corrosion products were formed before the coating was applied to the liner.

The area of maximum corrosion on liner PF-3 was determined to be the bare area on the inside surface where the coating had been removed intentionally for a conductivity probe. A section was removed from that area for metallurgical evaluation (Fig. 7). Corrosion in that area was uniform, with no evidence of pitting or pitting-type corrosion (Fig. 8). Comparing the minimum measured wall thickness in that corroded area with the measured uncorroded base metal thickness indicates a calculated minimum lifetime for liner PF-3 of 50 years. That exceeds the required liner storage time of 13 years at INEL. The resulting information was used to assess the ability of the liners to contain the highly radioactive contents during storage at INEL and to assist in selecting materials for high integrity containers for final disposal of the EPICOR-II prefilters. Work on this project is complete.

FUTURE PLANS

The following subsections describe plans for future work under this program.

Task 1--Resin Degradation

During early FY-1986, the second sampling of resins from the study prefilters (PF-8 and -20) was accomplished and the samples were analyzed chemically and physically to measure additional degradation. The NUREG report comparing analyses of the first and second samplings will be completed this fiscal year.

The chemical and physical analysis of resins from the third sampling will be completed in FY-1988. A topical report will be prepared comparing results of the first three analyses. The fourth resin sampling and sample analysis will occur in FY-1991, and a topical report will be written that year. The fifth resin sampling and analysis will occur in FY-1994. The fifth resin sampling will include collecting materials from two control prefilters (PF-9 and -27) stored with the study prefilters. Analysis of those samples will be completed that same year. Results of all the resin degradation analyses will be integrated and compared

[a]GEND is the acronym for General Public Utilities Nuclear Corporation, Electric Power Research Institute, U.S. Nuclear Regulatory Commission, and U.S. Department of Energy.

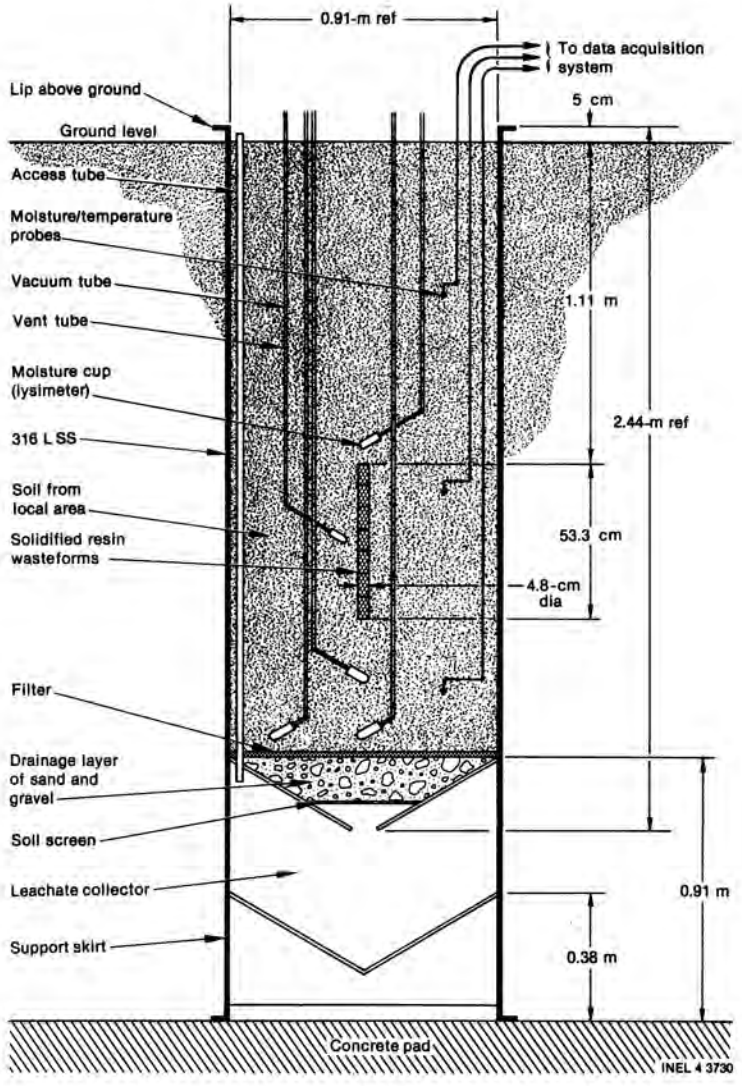


Fig. 6. Full Section Schematic of a Lysimeter Used for Field Testing EPICOR-II Resin Waste Forms.

Fig. 7. Metallographic Section Removed From the wall of Liner PF-3, Showing Corroded Interior Surface.

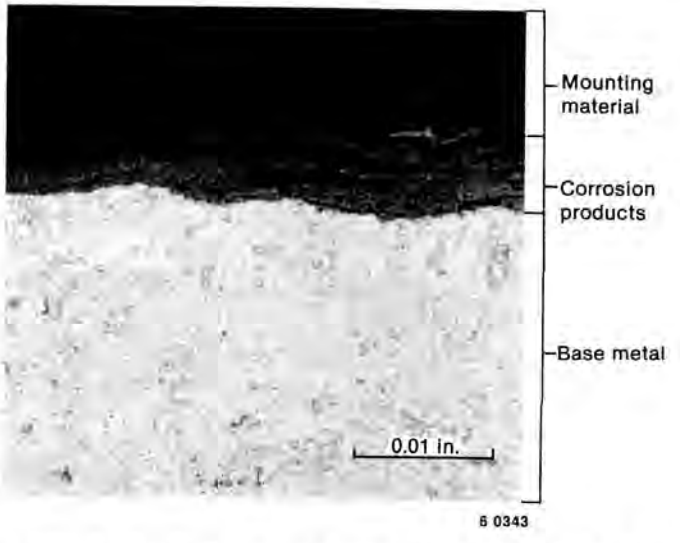


Fig. 8. Photomicrograph Normal to the Interior Surface of Section 1 Taken From Prefilter PF-3, showing the Uniformly Corroded Interior Surface.

to ascertain the rate of resin degradation versus time as a function of cumulative radiation dose. It is expected that degradation will progress with time and that a significant amount will have occurred by the end of 10 years. A final NUREG report on the task will be written and issued in FY-1994.

Task 2--Resin Solidification

Examination of biodegradability of the waste forms will be completed in FY-1986, and a topical report written and released, completing work on this task. EG&G Idaho will evaluate the test methods recommended in the TP in terms of their use for demonstrating stability of ion exchange media wastes. Some alternative testing protocols will be recommended.

Task 3--Field Testing

Field testing experiments will continue through FY-1986. A letter report will be issued describing the previous year's results, and a topical report is being written describing the experiment.

The experiments will be operated and monitored on a continuing basis through FY-2004, a total of 20 years. Letter reports will be issued annually, presenting the results of measurements to date. A topical report will be written presenting interpreted results and comparisons with results from other lysimeter experiments and leach tests of Task 2. That report will be issued at the end of the project.

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