PROCESSING AND REMOVAL OF THE THREE MILE ISLAND MAKEUP AND PURIFICATION SYSTEM RESINS

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ABSTRACT

The accident at Three Mile Island Unit 2 in March 1979 left the makeup and purification demineralizers inaccessible to workers. These demineralizers contained the highest concentrations of radioactive isotopes outside the Reactor Building. Additionally, unknown quantities of fuel were in the demineralizers. Other concerns centered on the unknown condition of the demineralizer resins, the possible loss of system integrity, and the presence of hydrogen due to the radiolytic breakdown of water contained in the demineralizers. GPU Nuclear, with the U.S. Department of Energy and its contractors EG&G Idaho, Inc., Oak Ridge National Laboratory, and Westinghouse Hanford Company, remotely measured the fuel and fission product inventory of the demineralizers and demonstrated a practical chemical processing method using samples acquired from the demineralizers. Equipment was developed and installed to elute the radioactive cesium from the demineralizers. And elution, which began in September 1984, now nears completion.

INTRODUCTION

During the Three Mile Island Unit 2 (TMI-2) accident, the resins in the A and B tanks of the makeup and purification demineralizer system were highly contaminated by the letdown of reactor coolant through the system. Large amounts of fission products, including cesium, strontium, and iodine, and fuel debris were trapped by the organic resin, resulting in resin bed temperatures estimated as high as 810 K from fission product decay heat and reactor coolant water. Radiation exposure to the resins was estimated to exceed 10^9 rads. Tank A was estimated to contain as high as 3500 Ci and tank B as high as 7500 Ci of radioactive cesium. Because of this high radioactivity, the resins and vessels had to be remotely characterized.

CHARACTERIZATION PROGRAM

The demineralizer cubicles were surveyed for physical damage using closed circuit television cameras mounted on a remote-controlled robot called SISI (for Surveillance and Inservice Inspection). The robot was modified for this project by Westinghouse Hanford Company. SISI performed radiation surveys. Figure 1 presents the results of this survey for cubicle B. Swipes for contamination also were obtained and later analyzed. The tanks were in good physical condition, with no evidence of leakage, but radiation fields around the tanks were in the hundreds of roentgens per hour and up to several thousand roentgens per hour at the tanks' surfaces.

Cameras on SISI helped workers to remotely place radiation monitoring instruments in cubicle A. The instruments were lowered into the cubicle through wall penetration #891 (see Fig. 2). An ion chamber detector and a ^{235}U neutron detector were positioned at opposite sides and across the top of demineralizer A to obtain gross gamma and neutron dose rates. The gross gamma dose rates were as high as 2780 R/h next to the tank, as shown in Fig. 3, but useful gross neutron data were unobtainable. Personnel could not develop similar radiation profiles for demineralizer B because its cubicle walls do not have an accessible penetration.

Other special equipment was adapted to determine fuel quantities in the demineralizers. Fuel estimates were obtained using a Compton recoil gamma spectrometer that was remotely positioned around the A tank, a beryllium (gamma, neutron) detector that was rolled in on a cart through the cubicles' doorways, and solid state track recorders that were lowered through penetration #891 and remotely positioned against the side of the A tank. All these systems recorded less than 4 kg of fuel in the two demineralizers and eliminated concerns regarding criticality; 70 kg per demineralizer would be required for criticality under optimal conditions. The instruments did, however, identify a transuranic specific activity in the resins higher than allowed at a commercial disposal site.

The results of the characterization efforts revealed that demineralizer B had as high as 7500 Ci of ^{137}Cs and less than 1 kg of fuel. Demineralizer A had as high as 3500 Ci of ^{137}Cs and an estimated fuel content of less than 2.5 kg.

Operations personnel sampled the gas in the tanks to determine hydrogen and oxygen concentrations. The gas sampling panel that was modified for the work also permitted purging and inerting of the internal atmosphere with nitrogen. This operation eliminated the concern about the formation of potentially flammable gas mixtures from radiolysis and, in

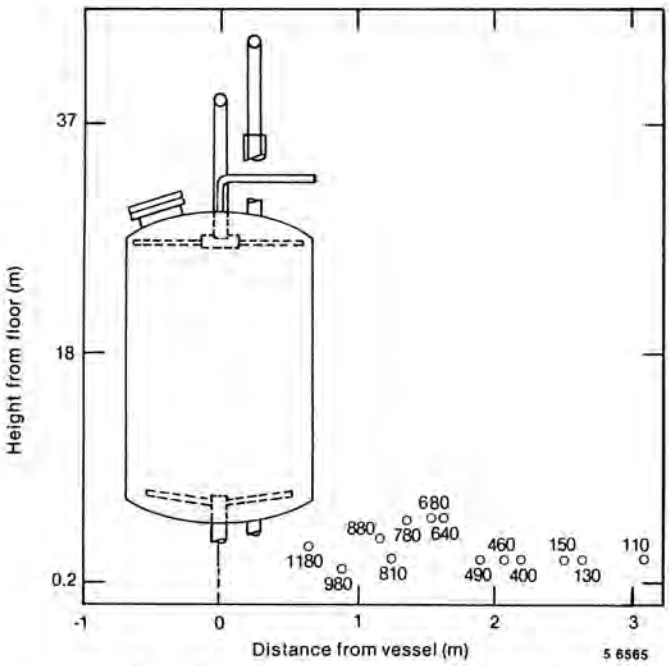

Fig. 1. Gamma dose rates (R) in cubicle B.

conjunction with a monitoring program, placed the demineralizers in a safe condition. In an analysis of the gas contents, examiners found high concentrations of H_2 in both demineralizer tanks (81% in tank B) but no O_2 in either tank.

The next step in characterizing the tanks' contents was to ascertain the physical condition of the resins and obtain actual samples. Because the radiation fields were as high as 2780 R/h at the demineralizer vessels, certain piping pathways could not be used for sampling, especially given the unknown condition of system components and the potential loss of system integrity that might have resulted with the operation of valves that had not been used or maintained for several years.

GPU Nuclear selected the resin fill lines, shown in Fig. 2, as the access points for sampling the resin beds. The resin fill lines are 7.6-cm, schedule-40 pipes that run from the Auxiliary Building corridor, through the gas analyzer room, and through penetrations into the adjacent demineralizer cubicles,

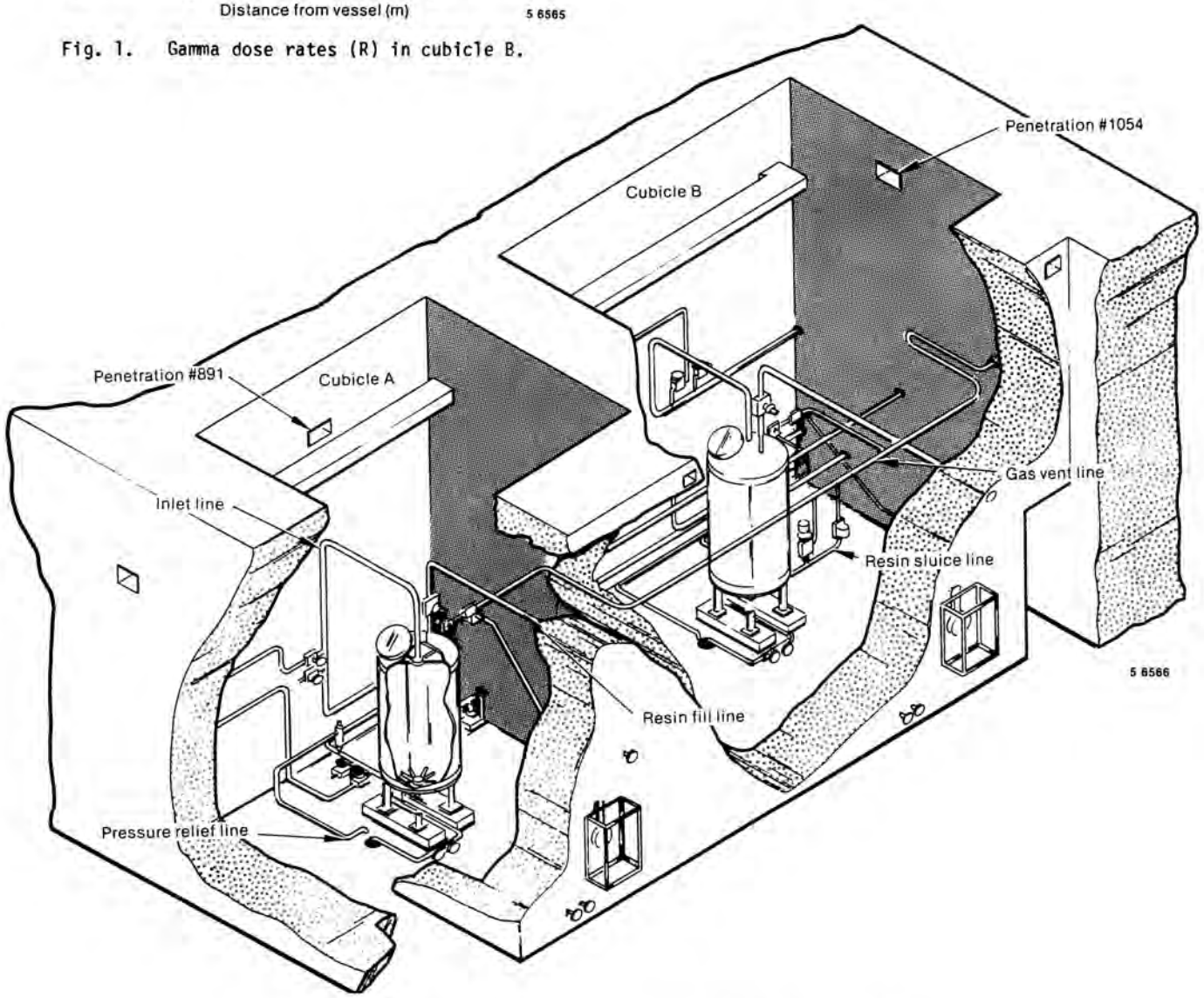

Fig. 2. TMI-2 makeup and purification demineralizer cubicles.

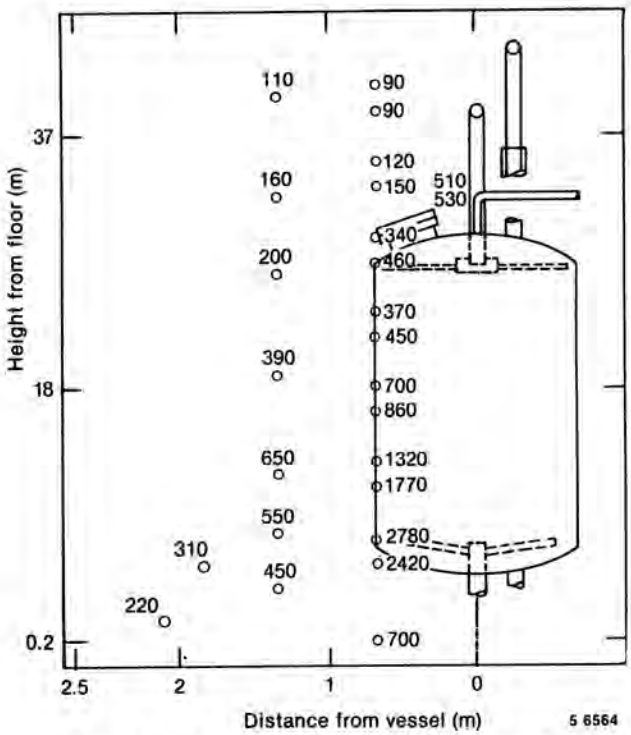

Fig. 3. Gamma dose rates (R) in cubicle A.

joining the demineralizer vessels on their upper pressure heads. The fill lines are isolated by 7.6-cm, air-to-open, spring-to-close, weir diaphram valves that are located in the demineralizer cubicles. These isolation valves presented a problem for sampling because the sample retrieval tool had to be flexible enough to pass over the weir to pass through the valve. Mockups of the valve piping system demonstrated that flexible 1.3-cm-inside-diameter plastic tubing would meet this need. The resin fill lines were cut just outside the cubicle walls in the gas analyzer room and fitted with flanges and valves to provide secondary isolation and equipment interfacing capabilities.

GPU Nuclear designed special sampling tools capable of entering each vessel through the piping and valve system. If the pneumatic operator on the weir valve had failed, workers would have been able to manually jack the diaphragm valve open against its 907-kg spring.

Samples from each of the resin beds were manually obtained from approximately 7.6 m away using this remotely operated tooling. From demineralizer B, workers acquired a liquid and solid sample; the resin bed was covered with water. Fiberoptic observations of demineralizer A revealed that the resin bed was dry, with a void in the middle of it, and covered with a crust, preventing technicians from obtaining more than a few grams of solid material. The A demineralizer was resampled after water was added and the resin bed was nitrogen sparged to resuspend the solids; technicians acquired a sample of liquid and solid. The samples all were analyzed in laboratory hot cells.

Subsequent to this analysis, several methods of removing the irradiated resin were studied. All of the concepts included an elution process to rinse the cesium from the resins because of the high level of radiation. Elution would lower the activity in the resins before their removal from the demineralizers.

The basic approach to each concept was similar: a chemical solution would be used to rinse the resins, which would then be sluiced out of the existing 6.4-cm resin sluice line. All of the concepts called for the use of plant processing water and a combination of existing and new piping. The methods differed in the types of equipment used, the tank for storing the sluiced resins before shipment, and the location of the system in the Auxiliary Building. GPU Nuclear decided that the resins would be sluiced into the spent resin storage tank.

TMI-2 PLANT DEMINERALIZER SAMPLE ANALYSIS

Oak Ridge National Laboratory (ORNL) analyzed the samples from demineralizer vessels A and B to validate GPU Nuclear's plans to reduce the dose levels in the demineralizer resins by rinsing and chemically processing them. In a laboratory simulation, ORNL scientists subjected sample materials from both demineralizer vessels to a 17-stage rinse and elution process. In the initial stage of the process, sample material was rinsed with 0.18 molar boric acid. In subsequent stages, the ion-exchange resin samples were subjected to increasing concentrations of sodium hydroxide. Figures 4 and 5 present the results of the simulated cesium elution process for the A and B resin samples, respectively. The test results indicate that 90% of the cesium could be eluted from the B demineralizer sample, and about 70% could be eluted from the A demineralizer sample. The specific activity of the A and B samples was similar after processing. Also during the test, scientists examined how effectively the zeolite ion-exchange material in the TMI-2 Submerged Demineralizer System (SDS) could

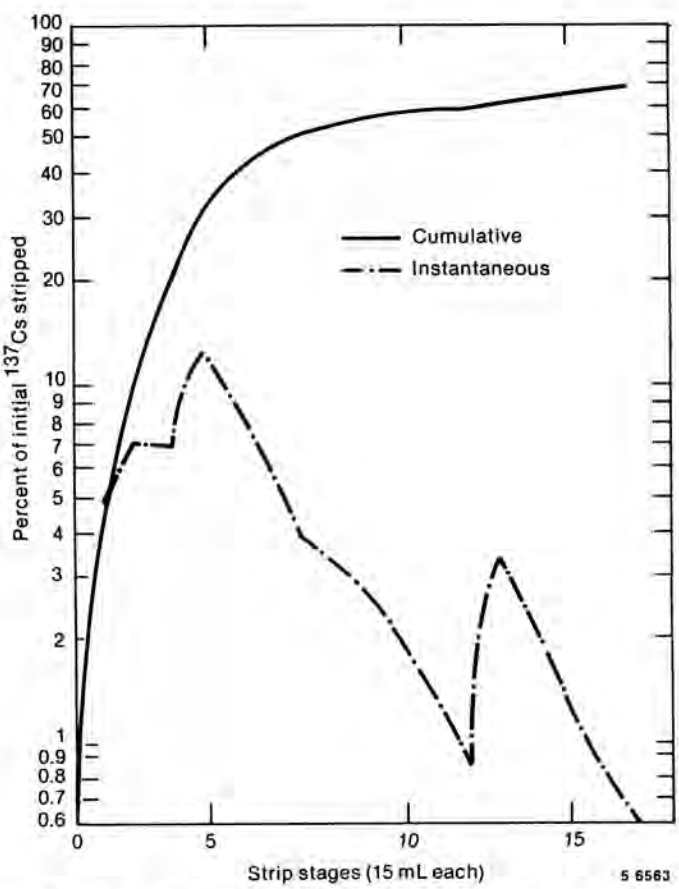

Fig. 4. Stripping of ^{137}Cs from a bed of column A resin by successive batch contacts.

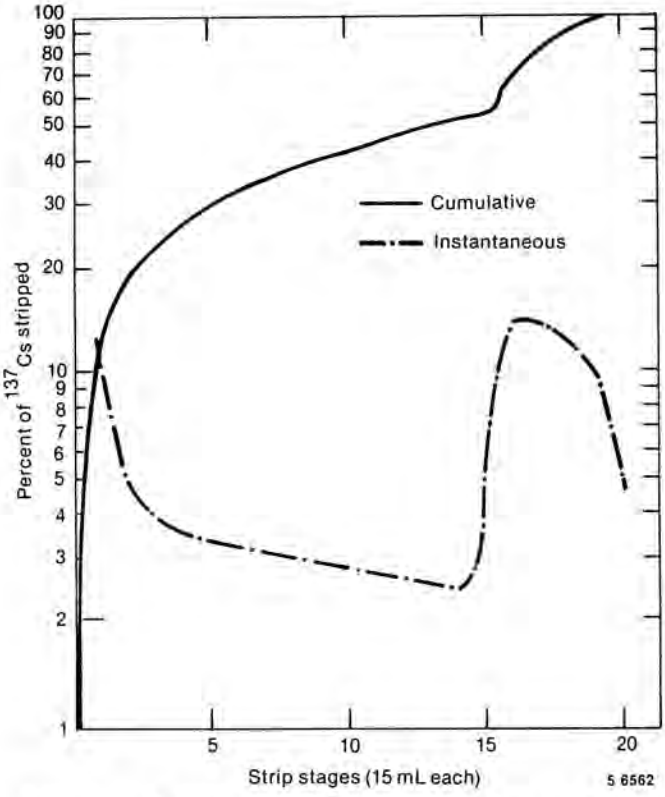

Fig. 5. Stripping of ^{137}Cs from a bed of B-2 resin by successive batch contacts.

remove cesium from the eluant. Results documented that the SDS system could remove 99.99% of the cesium.

ORNL scientists also studied GPU Nuclear's plans for filtration of effluent before SDS treatment. Filtering was intended to prevent the unnecessary introduction of particulate matter to the SDS ion-exchange materials. ORNL's filtering tests showed that a small-pore (0.5×10^{-6} m), sintered, stainless steel filter could be quickly and irreversibly clogged by the process effluent, while larger-pore (10×10^{-6} m and greater), sintered, stainless steel filters could remove the particulate without the rapid plugging experienced by the filters with smaller pores.

In addition to the laboratory simulations described above, ORNL performed microscopic and macrophotographic examinations of the A and B demineralizer samples. These examinations indicated that even though the B sample was more highly loaded with fission products, the resin in the A sample had sustained more extensive physical damage. The B sample appeared to be somewhat discolored, yet the resin beads were mostly intact, and they had experienced no apparent charring or agglomeration. The A sample, however, experienced significant charring and bead agglomeration. Previous resin degradation experimental work by Battelle Pacific Northwest Laboratory showed that temperatures of at least 810 K are required for this resin to char.

Concluding their work, ORNL scientists verified the effectiveness of GPU Nuclear's plans to elute the cesium from the demineralizer resins, filter the resulting effluent through sintered, stainless steel

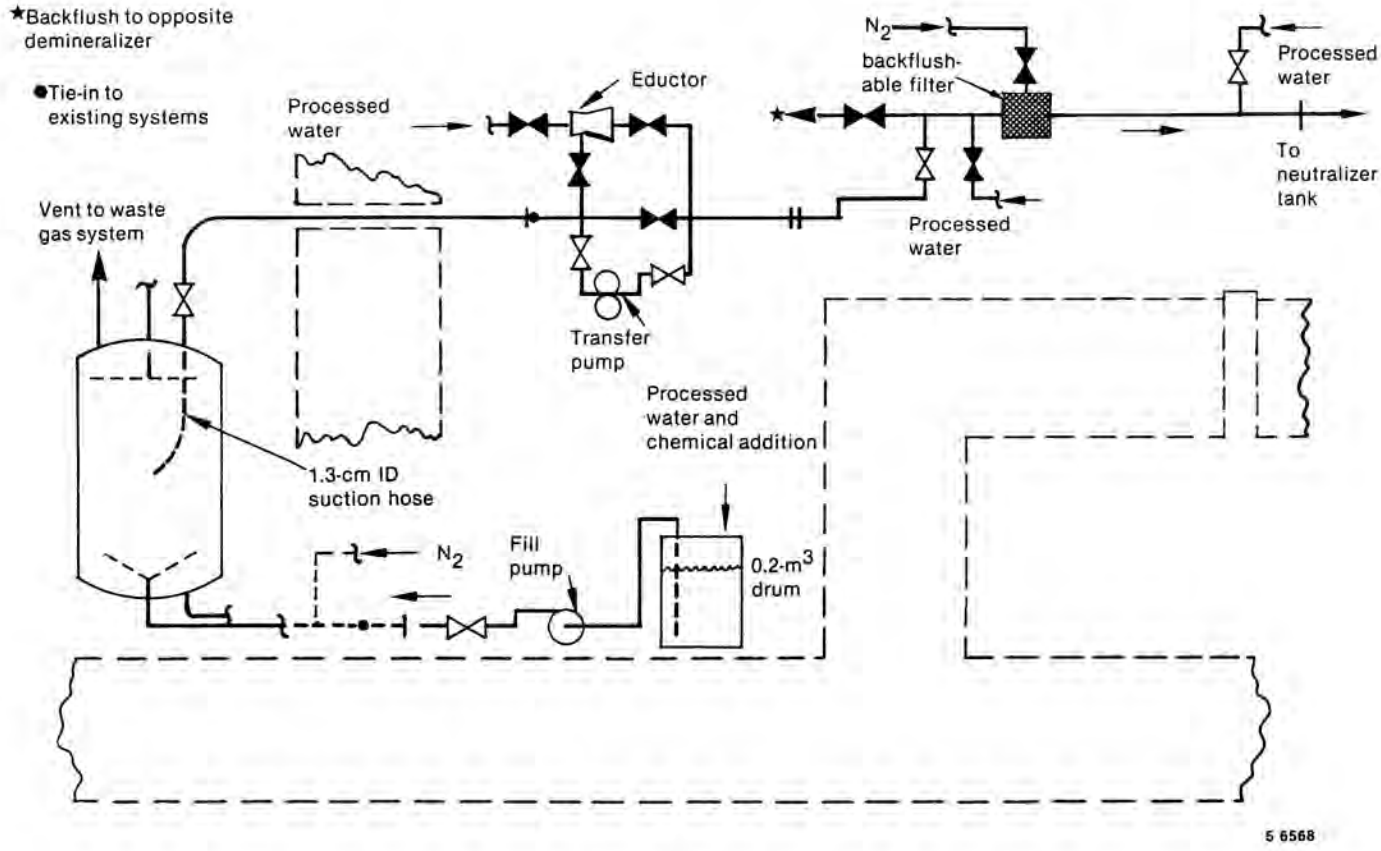

Fig. 6. Schematic for rinse and elution.

media, and use the SDS to remove the cesium from the eluant.

CESIUM ELUTION

Using the data gathered from the ORNL tests, Westinghouse Hanford Company designed, fabricated, and tested the elution and filtering system (see Fig. 6). A heavily shielded pump and eductor skid pumps water out of the demineralizer tank through a 1.3-cm-inside-diameter plastic tube that was fed through the resin fill line and weir diaphragm valve and inserted into the vessel. A chemical addition skid mixes water with chemicals and adds a batch of the solution to the demineralizer tank through the normal tank effluent line. Chemical concentrations can be adjusted in the chemical addition tank before each batch. The pump and eductor skid is designed to draw liquid from the demineralizer at 1.6×10^{-5} to 19×10^{-5} m^3/s and to delute the water 20:1 so that the eluant can be processed without harming the existing equipment downstream. The pump and eductor skid then delivers the diluted effluent to a filtration system located in the gas analyzer room 6.1 m away. This filtration system uses a 20×10^{-6} m, sintered, stainless steel filter that prevents suspended fuel particles and resin debris from reaching downstream equipment. When the pressure drop across the filter reaches 103,421 Pa, the filter is backflushed with nitrogen gas. The effluent water--containing cesium, strontium, organic contaminants, and other radionuclides--is then stored in the neutralizer tanks and processed in batches by the SDS. The majority of the radioactivity has been trapped in one SDS liner that will be part of the Department of Energy-monitored retrievable burial demonstration program in Hanford, WA.

A robot nicknamed "Louie," also designed and fabricated by Westinghouse Hanford Company, will take radiation measurements in the cubicles to demonstrate the dose level reductions after the process is completed. Louie, in Fig. 7, is a remote-controlled vehicle with a manipulator arm and three closed circuit television cameras, one mounted on the arm and two on the body of the vehicle. The robot's transporter base is 48 cm wide and 70 cm long so the robot can fit through the small doorways to the cubicles. It can reach 69 to 234 cm from the floor, and it has a grip strength of 34 kg--more than adequate for carrying the Eberline radiation monitor into the cubicles.

PROJECT STATUS

In the last week of February 1985, approximately 3500 Ci of the cesium activity in the B demineralizer vessel and approximately 800 Ci of the activity in the A demineralizer vessel were removed. A program has been established to measure the radiation levels in the tanks, and calculations will be made to estimate the remaining cesium activity. Samples will be taken and analyzed if necessary. The goal of this project is to reduce the concentration of radioactive cesium from each vessel so the resins can be safely sluiced. Dose rates in downstream pipes were as high as 200 mR/hr.

The still transuranic-contaminated resins will be sluiced to the spent resin storage tank using existing equipment and then packaged for shipment. Radiation levels in the empty vessels will determine the approach for their final decontamination.

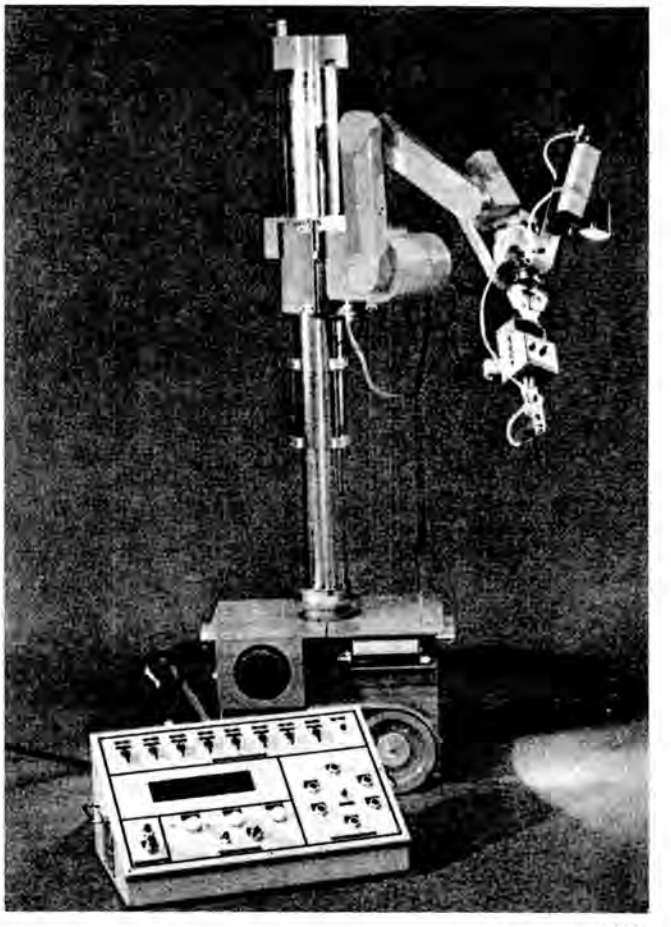

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Fig. 7. Remote-controlled "Louie" robot.