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Chemistry

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by
W. T. Withers
Pile Engineering Division

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E. I. du Pont de Nemours & Co.
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ABSTRACT

The ability of ion exchange resins to remove soluble neutron absorbers from the moderator of a reactor was investigated. The soluble absorbers are materials that might be used to poison a reactor for emergency shutdown. Each of the five poisons tested was completely removed from solution by ion exchange resins and none formed an insoluble film on aluminum surfaces.

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INTRODUCTION

The dispersal of neutron absorbers or poisons in the moderator has been proposed as a method for shutting down a reactor when other safety systems fail to operate in an emergency. For such a method to be practical, the poison should be readily removable from the moderator.

It is conceivable that some neutron absorbers would interact with the metal surfaces inside the reactor, leaving an insoluble layer of poison in the active lattice. Therefore, to insure adequate cleanup, the poison must not only be removed from solution but also must leave no surface residue.

The purpose of this study was to demonstrate on a laboratory scale that candidate poisons could be effectively removed from the moderator and from the aluminum components of a reactor by ion exchange.

SUMMARY

All of the poisons tested were readily removed from solution by ion exchange resins. None of the poisons left a detectable residue on aluminum. Candidate materials investigated were potassium tetraborate, cadmium nitrate, lithium borate, gadolinium nitrate, and mixed gadolinium-samarium nitrates.

DISCUSSION

In general, the experimental procedure to demonstrate removability of poisons and absence of surface contamination was as follows: Aluminum specimens were immersed in solutions of the poisons for about 50 hours. The poison solutions were then circulated through an ion exchange column until the dissolved salts were removed. The metal specimens were left in the resulting deionized water for another 50 hours. Before and after this treatment, the danger coefficient of each metal specimen was measured in the Standard Pile* to detect any residual poisons.

PREPARATION OF POISON SOLUTIONS

Because of their high neutron absorption, solubility in water, stability, and availability, compounds of one or more of the following elements were recommended for consideration as injectable poisons: gadolinium, samarium, cadmium, boron, and lithium. The compounds that were selected for study are:

Potassium borate, C.P. grade, $K_2B_4O_7 \cdot 5H_2O$, obtained from Fisher Scientific Company.

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* The Savannah River version of the General Electric Thermal Test Pile (Ref.1)

Cadmium nitrate, reagent grade, $Cd(NO_3)_2 \cdot 4H_2O$, obtained from the General Chemical Division of Allied Chemical and Dye Corporation.

Lithium borate, technical grade, $LiBO_2 . H_2O$, obtained from Foote Mineral Company. No information was available concerning the purity of this material.

Gadolinium nitrate 98%, Code 948, $Gd(NO_3)_3 . xH_2O$, obtained from Lindsay Chemical Company. On an oxide basis, this material contains 98% Gd_2O_3 and the remaining 2% is Sm_2O_3, Nd_2O_3, Dy_2O_3, and other rare-earth oxides.

Mixed samarium-gadolinium nitrates, code 940, $(Sm,Gd)(NO_3)_3 . xH_2O$, obtained from Lindsay Chemical Company. The manufacturer gives the following approximate oxide composition of this material:

Sm_2O_3	50-55%
Gd_2O_3	25-30%
Nd_2O_3	1-5%
Pr_6O_{11}	1-4%
CeO_2	0-1%
Eu_2O_3	1-1.5%
Other rare-earth oxides plus Y_2O_3	9-16.5%

Each poison was made up into a solution containing a total calculated neutron absorption cross section of approximately 1.4 cm^2/liter. The concentrations of these solutions are given in the second column of Table I. These concentrations were calculated from the formulae given above and the stated purity, when available. In the case of the lithium borate, where the purity is not known, no allowance for impurities was made. Both rare-earth salts were assumed to be hexahydrates. In addition, the absorption of neutrons by the 2% impurities in the gadolinium nitrate was neglected. For the mixed rare earths, a composition within the range given by the manufacturer was assumed.

PREPARATION OF METAL SPECIMENS

For early stages of the program, the metal specimens were half cylinders cut from a thin-walled tube of 63S aluminum 1.4 inches in diameter. The specimens were 5-3/8 inches long and had a total surface area of about 22 square inches. The dimensions were limited by the size of the largest opening then available in the Standard Pile. The tube from which these specimens were cut was one which had been stored under water for several weeks. This was done in an attempt to simulate the surface oxidation typical of that found in a reactor.

For the tests with the mixed gadolinium-samarium nitrates, a larger opening in the Standard Pile was made available. This opening permitted the use of cylindrical specimens two inches in diameter and ten inches long. These specimens were formed from 63S aluminum sheet stock and had a total area of 126 square inches. This metal had not been stored under water for any extended period, but the increased sensitivity for detection of poisons associated with the larger area was considered more important than slight differences in the surface.

DETAILS OF EQUIPMENT

The laboratory equipment for these experiments consisted of a 3-liter battery jar with tubing fittings on the side and bottom. Connected to these fittings was a clean-up loop containing a rotameter, an ion exchange column, a flow-type conductivity cell, and a small centrifugal pump arranged as shown in Figure 1. The ion exchange column was 0.5 inch in diameter and was filled to a height of 24 inches with a mixture of "Amberlite" resins IR-120 and IRA-400. Appropriate pinchcocks were provided in the connecting lines to control the flow and to isolate the loop from the storage vessel.

DETAILS OF PROCEDURE

Each aluminum specimen was placed in the Standard Pile and the danger coefficient of the specimen was measured.

Each specimen was then immersed in a solution of one of the candidate poisons contained in the storage vessel described earlier. After an immersion period of about 50 hours, the poison solution was pumped through the ion exchange bed at a flow rate of 100-140 cc/min. The resistivity of the water leaving the resin bed was measured to determine the effectiveness of the bed in removing the poisons. The resistivity of the bulk solution in the storage vessel was also measured with a dip-type conductivity cell.

After the resistivity of the water in the open storage vessel had leveled off at above 5×10^6 ohm-cm, the circulating pump was shut off. The specimens were then left in the deionized water for about 50 hours. At the end of this clean-up period, the specimens were removed and dried in air. The entire operation was carried out at room temperature.

Each specimen was again placed in the Standard Pile and the danger coefficient was measured. The change in danger coefficient, if any, was attributed to residual poison and was reported as cm^2 of neutron absorber.

RESULTS AND CONCLUSIONS

The results of the immersion tests are presented in Table II. In all cases, the specific resistivity of the water leaving the resin bed was high, 12.0×10^6 to 17.5×10^6 ohm-cm. This indicated complete removal of the poisons from solution. As is to be expected with a resin system that removes all the ionic matter passing through, the time required for cleaning up the bulk water depends directly on the initial concentration and inversely on the flow rate.

The measured absorptions attributed to residual poisons are given in the last column of Table I. Each sample had a total residual poison of less than 11.0×10^{-3} cm^2, which is the estimated limit of sensitivity for the Standard Pile. It was therefore concluded that the poisons tested do not leave a residual film on aluminum within the sensitivity of the technique. For the specimens that had a surface area of 22 in^2, the sensitivity limit corresponds to a maximum of 5×10^{-4} cm^2 of absorber/in^2 of surface. The sensitivity limit for the 126 in^2 specimen is 8.7×10^{-5} cm^2 of absorber/in^2 of surface.

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FIGURE 1

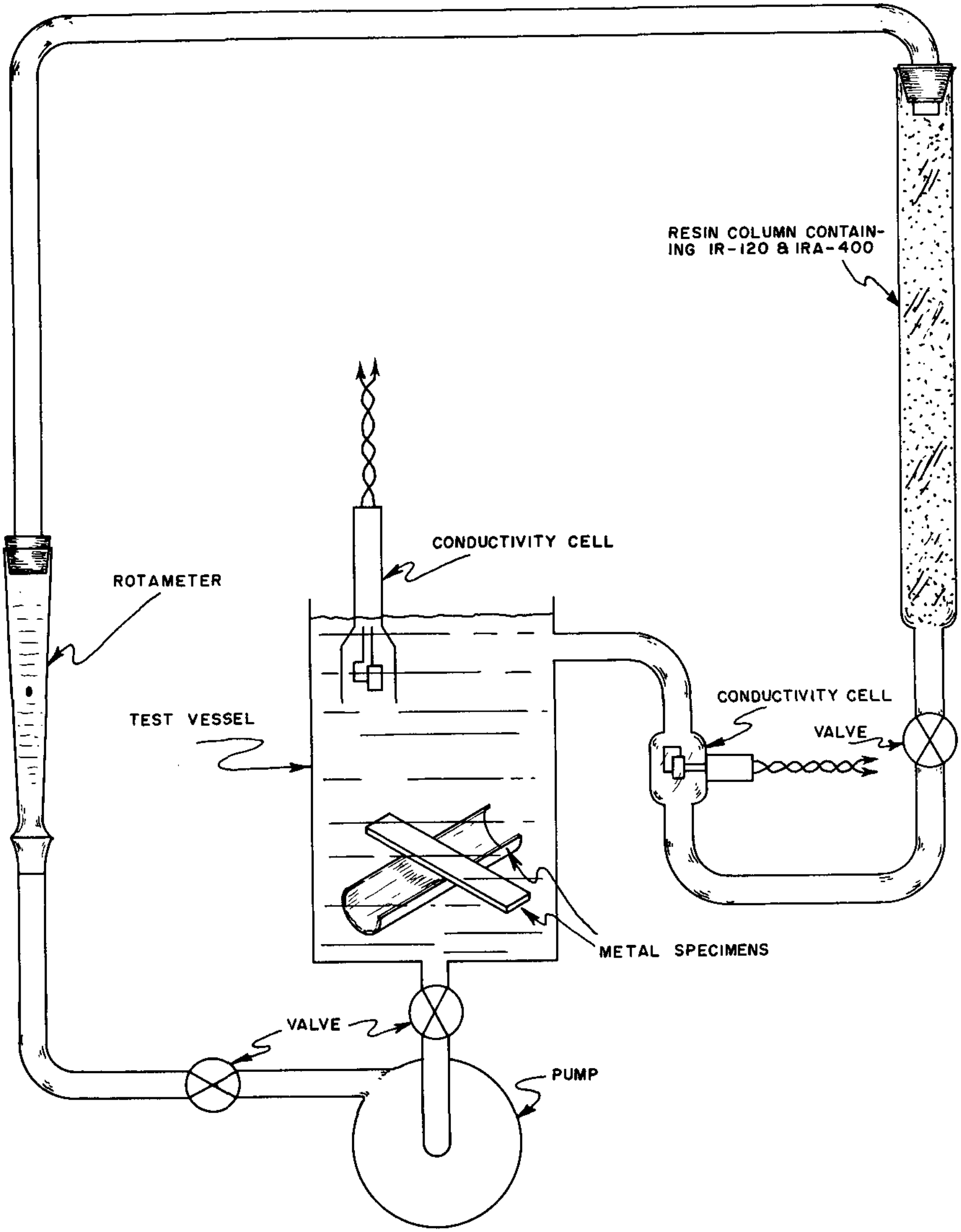

DIAGRAM OF CLEAN-UP LOOP

TABLE I

RESULTS OF IMMERSION TESTS

Absorber Tested	Concentration of Absorber, grams/liter	Area of Specimen, in^2	Residual Poison, cm^2
$Cd(NO_3)_2 \cdot 4H_2O$	0.23	22	-8.6×10^{-3}
$K_2B_4O_7 \cdot 5H_2O$	0.266	22	$+3.4 \times 10^{-3}$ (a)
$Gd(NO_3)_3 \cdot 5H_2O$	0.0288	22	$+3.4 \times 10^{-3}$
$LiBO_2 \cdot H_2O$	0.204	22	$+10.0 \times 10^{-3}$
Mixed Gd-Sm Nitrates	0.069	126	$+1.0 \times 10^{-3}$

(a) Note: A black deposit formed on scarred areas of the sample during immersion but was removed upon cleanup.

TABLE II

DATA ON PERFORMANCE OF TEST LOOP IN REMOVING POISONS

	TEST OF CADMIUM NITRATE	TEST OF POTASSIUM TETRABORATE
Clean-up Time, Min	Specific Resistivity of Water in Vessel, ohm-cm	Specific Resistivity of Water in Vessel, ohm-cm
10	8.0×10^3	8.0×10^3
30	24.0×10^3	21.0×10^3
45	47.0×10^3	26.0×10^3
85	360×10^3	170×10^3
----------- Run interrupted for 48 hours at this point--------------		
95	600×10^3	345×10^3
115	800×10^3	590×10^3
150	2.4×10^6	2.4×10^6
260	5.5×10^6	5.8×10^6

Note: Loop flow rate for above tests = 100 ml/min

(Continued on page 10)

TABLE II (Continued)

DATA ON PERFORMANCE OF TEST LOOP IN REMOVING POISONS

	TEST OF LITHIUM BORATE	TEST OF GADOLINIUM NITRATE
Clean-up Time, Min	Specific Resistivity of Water in Vessel, ohm-cm	Specific Resistivity of Water in Vessel, ohm-cm
0	5.5×10^3	37.0×10^3
10	7.6×10^3	65.0×10^3
20	17.5×10^3	105×10^3
50	72.0×10^3	590×10^3
80	350×10^3	900×10^3
105	1.0×10^6	4.6×10^6
170	8.7×10^6	8.0×10^6
205	8.7×10^6	8.5×10^6

Note: Loop flow rate for the above tests = 140 ml/min

TEST OF MIXED GADOLINIUM-SAMARIUM NITRATE

Clean-up Time Min,	Specific Resistivity of Water in vessel, ohm-cm
0	13×10^3
40	1.3×10^6
45	2.4×10^6
60	5.2×10^6

Note: Loop flow rate was not measured in this test