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LEACHABILITY OF DECONTAMINATION REAGENTS FROM CEMENT WASTE FORMS.*

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Brookhaven National Laboratory, in order to provide technical information needed by the U.S. Nuclear Regulatory Commission to evaluate the adequacy of near-surface disposal of decontamination wastes, has begun to study the leachability of organic reagents from solidified simulated decontamination wastes. Laboratory scale cement waste forms containing EDTA, picolinic acid or simulated LOMI decontamination reagent were leach tested. Samples containing an organic reagent on either mixed bed ion-exchange resins or anion exchange resins were tested. A fixed interval leach procedure was used, as well as the standard procedure ANS 16.1. The leachability indices measured for the release of the acid from resin/cement composites are: 10.1 for EDTA on mixed bed resins; 9.1 for picolinic acid on mixed bed resins; 9.2 for picolinic acid on anion exchange resins; 8.8 for picolinic acid in forms containing simulated LOMI reagent on mixed bed resins and 8.7 for picolinic acid in forms containing simulated LOMI reagent on anion exchange resins. The leachability indices measured varied with leach time and the data indicate that the release mechanism may not be simply diffusion controlled.

*Work carried out under the auspices of the U.S. Nuclear Regulatory Commission.

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Light water reactors may require one or more chemical decontaminations to achieve their designed lifetimes. Primary system decontamination is designed to lower radiation fields in areas where plant maintenance personnel must work. Commercially available chemical decontamination processes contain organic acids and chelates. These organic reagents are then present in the subsequent radwaste and if released to a disposal site, may enhance the migration of radionuclides by their ability to form soluble complexes with the radionuclides.

Brookhaven National Laboratory has begun to study the leachability of organic reagents from cement solidified decontamination wastes in order to provide technical information needed by the U.S. Nuclear Regulatory Commission to evaluate the adequacy of near-surface disposal of decontamination wastes. Laboratory scale cement waste forms containing EDTA or picolinic acid on organic ion-exchange resins were leach-tested, and a leachability index for the organic acid was calculated according to the procedure given in the draft standard, ANS 16.1.(1)

Five different decontamination reagent/resin combinations solidified in Portland I cement were leach tested. All of the laboratory scale waste forms were a nominal 2-in. diameter by 4-in. long right cylindrical solid. Details of the solidification methods are given elsewhere.(2,3) The decontamination reagents tested were: EDTA, picolinic acid, and a simulated LOMI** reagent. The LOMI reagent was simulated with an equimolar mixture of picolinic acid and formic acid. The compositions of the samples tested are summarized in Table 1.

Table 1

Composition of Simulated Decontamination Waste Leach Samples

Sample ID	Reagent	Resin Type
EDTA/MB	EDTA	IRN-77/IRN-78
PIC/MB	Picolinic acid	IRN-77/IRN-78
LOMI/MB	LOMI	IRN-77/IONAC A-365
PIC/AN	Picolinic acid	IRN-78
LOMI/AN	LOMI	IONAC A-365

*Work carried out under the auspices of the U.S. Nuclear Regulatory Commission.
**Low oxidation metallic ion.(4)

Three different schedules for changing the leach solutions were used. In a fixed increment test, the first leachate change interval was 8 days and then every 7 days for a total of 92 days. This procedure was used for samples EDTA/MB and PIC/MB. After finding that the quantities of organic acid in the leachate were detectable with the analytical methods available, the test schedule was changed to one outlined in the draft ANS 16.1 procedure. However, two slightly different schedules for changing the leachate were used. Both ANS procedures started with a 30-second rinse of the form followed by leachate changes at 2 h, 7 h, 24 h, 48 h, 72 h, and 96 h. Then one schedule used for sample LOMI/MB was based on the November 1982 draft of ANS 16.1 in which the leachate was changed every 7 days to a total of 95 days. The other samples, LOMI/AN and PIC/AN, had additional leachate changes at 120 h, 19 d, 47 d and 89 d. Methods for the analysis of the leachates for EDTA or picolinic acid can be found in Piciulo et al.[2]

The leachability indices for the release of the organic acid from the waste forms are summarized in Table 2. The value given is the average of all the leach indices determined during the test. The percent bias given in the table indicates that the average of the first four leach indices is that percentage larger (+ sign) or smaller (- sign) than the average of the last four leach indices. The ranges of the leach indices are also given with the range percent in parenthesis. In all cases, the cumulative fraction leached from the samples was less than 20% of the quantity of the organic acid initially present in the form and thus the effective diffusivity and leachability index were calculated using the equations given in ANS 16.1.

Table 2

Summary of Samples Tested and Leach Index Calculated

Sample ID	Acid Measured	Leach Index	Leach Index Range
EDTA/MB	EDTA	10.1 +1%	9.8 - 10.3 (5%)
		10.1 +1%	9.7 - 10.3 (6%)
PIC/MB	Picolinic Acid	9.1 +2%	8.8 - 9.2 (5%)
		9.1 +3%	8.8 - 9.2 (4%)
LOMI/MB	Picolinic Acid	8.8 +8%	8.3 - 9.1 (9%)
		8.8 +7%	8.3 - 9.1 (9%)
		8.8 +8%	8.3 - 9.1 (8%)
PIC/AN	Picolinic Acid	9.2 -3%	8.9 - 9.6 (7%)
		9.2 -3%	8.9 - 9.6 (7%)
		9.2 -3%	8.9 - 9.6 (7%)
LOMI/AN	Picolinic Acid	8.7 -2%	8.6 - 8.8 (3%)
		8.1 -5%[a]	7.8 - 8.6 (12%)
		8.7 -1%	8.6 - 8.8 (2%)

[a]Waste form fractured after leachate change at 7 hours.

The bias for the leach index values suggest a small increase or decrease as leach time progresses. Close examination of the leach index vs leach time showed regions of distinctively different leach index values. During the first 20 days of the ANS 16.1 test, there is a gradual increase in the leach index for the release of picolinic acid from LOMI/MB samples. After 40 days, a constant average leach index was reached and maintained throughout the remainder of the test. A different variation in leach index with progressing leach time was observed for the release of picolinic acid from PIC/AN samples which contain the acid on IRN-78 anion exchange resins. In this case, the leach index decreased during the first week of leachate sampling and then increased during the remainder of the test period.

The leachability indices measured for EDTA and picolinic acid were greater than the value of six which is the recommended minimum acceptable leach index for radionuclides given in the Technical Position on Waste Form.[5] This initial data indicated that the leach index is specific for the acid in the form. Further the release of an acid from a form appeared to be affected by the type of resin in the form. Although the leach index increases with time (release of acid decreases), which may be a desirable behavior, the data indicate that the release mechanism may not be simply diffusion controlled. Uncertainty in the release mechanism can make an extrapolation of the leach index to large forms misleading. In turn, source terms calculated for modeling efforts may be poorly defined.

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