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Permissible Radionuclide Loading for Organic Ion Exchange Resins from Nuclear Power Plants

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ABSTRACT

A questionnaire on the use of ion exchange resins in nuclear power plants was sent to all operating reactors in the U.S. Responses were received from 23 of the 48 utilities approached. Information was sought concerning the amounts of radionuclides held by the resins, and the effects of its radiation on the resins both during operation and after removal from service. Relevant information from the questionnaires is summarized and discussed. Available literature on the effects of ionizing radiation on organic ion exchange resins has been reviewed. On the basis of published data on damage to resins by radiation, the technical rationale is given to support NRC's draft branch technical position on a maximum permissible radionuclide loading. It is considered advisable to formulate the rule in terms of a delivered dose rather than a curie loading. A maximum permissible dose of 10^8 rad is chosen because, while it is large enough that a measurable amount of damage will be done to the resin, it is small enough that the damage will be negligible at a power plant or disposal site. A test procedure has been written which a generator could use to qualify a specific resin for service at a higher dose than permitted by the general rule.

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PERMISSIBLE RADIONUCLIDE LOADING FOR
ORGANIC ION EXCHANGE RESINS FROM NUCLEAR POWER PLANTS

1. INTRODUCTION

Organic ion exchange resins are used in nuclear power plants to control the purity of such liquid streams as the primary coolant, turbine condensate, water in the spent fuel storage pools, and the liquid radwaste resulting from all phases of plant operation. In the process, the resins pick up radioactive ionic species and particulate matter. The amount of radioactivity can be relatively large, particularly for resins used to treat the primary coolant.

All organic molecules, including those constituting ion exchange resins, undergo decomposition when exposed to ionizing radiation. Therefore, the resins used in power plants could be significantly degraded if they picked up sufficient activity before being removed from service, since degradation can occur long after removal from service.

In order to obtain information regarding the magnitude of such radiation damage received in actual power plant practice, a questionnaire on resin usage was sent to all the operating light water power reactors in the U.S.

This questionnaire differed from those used in previous surveys[1,2] in focusing more on disposal methods and practices than on operating conditions and procedures. For purposes of the present report, information was sought particularly on the levels of radioactivity to which the resins are exposed during operation, and the curie loadings on the spent resins which are shipped for disposal. The results of the survey responses are reviewed in the next section of the report.

Information on the effects of ionizing radiation on ion exchange materials published up to about 1964 is given in a book by Egorov and Novikov.[3] The book covers most of the work done in Russia and Eastern Europe, but there are gaps in the coverage of papers published in the West. These gaps were filled and the subject brought up to date to approximately the end of 1977, in a comprehensive review by Gangwer, Goldstein and Pillay.[4] More recent data of particular interest in the evaluation of effects of high radiation doses to resins used for nuclear waste disposal have been obtained from work done in connection with the TMI-II auxiliary building (AFHB) cleanup activities. Several studies were done on resins similar to those employed at TMI, using high integral doses.[5-7] Results of these studies were reviewed in a recent BNL report[8] and will be discussed in Section 3.2.4.

There is wide variation in resistance to radiation of the many organic cation and anion exchange resins available commercially. However, there appears to be sufficient similarity in behavior among the relatively few different nuclear grade resins of each class (cation and anion) presently used in nuclear power plants that a reasonable assessment can be made of a maximum

acceptable radiation dose for these resins as a class. On the basis of the data that have been published, NRC has recommended in a draft branch technical position[9] that resins should not be loaded to bulk specific activities greater than 10 Ci/ft^3, which is approximately equivalent to a dose of 10^8 rad for long-term decay of Cs-137 and Sr-90. In the event that the waste generator wishes to load resins to a higher level than 10 Ci/ft^3 (and thus subject them to a long-term dose greater than 10^8 rad) NRC would require demonstration that the resin proposed for use will not undergo significant radiation degradation at the higher loading.

The purpose of this report is twofold. First, it presents the technical rationale in support of the maximum loading limit. Second, since the maximum loading criterion is to a certain extent based on generic analysis of radiation effects on organic ion exchange media, an attempt is made to define a test for use on a case-by-case basis to determine whether, for a given resin, a higher total dose than 10^8 rad might be acceptable. The discussion in this report relies mainly upon effects observed for unsolidified resins. Thus, by and large, the impact of solidification upon the maximum loading limit was felt to be outside the scope of this study. There is limited evidence which indicates that cement solidification of organic ion exchange media may mitigate the deleterious effects seen upon irradiation of unsolidified resins. If this indeed should prove to be the case, then a reasonable maximum acceptable loading limit for solidified resin waste may well be different than for unsolidified resin waste. Since insufficient data exist to make this assessment, however, recommendations for future work in this area have been included.

2. RESPONSES TO SURVEY QUESTIONNAIRE

The questionnaire on usage of ion exchange resins was sent to 48 U.S. utilities operating 65 nuclear power plants. Responses were returned by 23 companies, giving data for 34 units, and information was obtained by telephone from two other companies. A copy of the questionnaire is reproduced in Appendix A, along with tables summarizing the answers to questions which were of particular interest for this report. Copies of most of the completed questionnaires which were returned are included in an appendix to a report on high integrity containers* to be published soon.

2.1 Resin Use and Operating Conditions

Relevant information on these topics as gathered from the survey responses is summarized below.

2.1.1 Kinds of Resin

(a) It appears that inorganic ion exchangers are not used in commercial nuclear power plants on a routine basis. All responses to the survey question on use of inorganic resins were negative.

(b) The most frequently used organic resins were strong acid cation exchangers (with sulfonic acid functional groups), and strong base anion exchangers (with quaternary ammonium functional groups). It appears to be the practice throughout the industry to use nuclear grade resins. A few plants did not report the specific resins used, but the remainder indicated use of nuclear grade only. (A description of nuclear grade resins is given in Section 3.2.5.) For the beds used to clean primary coolant (those of most interest to this report because of the higher radiation doses they receive), the majority of reactors use mixed bed resins, with cation: anion ratios usually 1:1, though a number use separate cation and/or anion beds as well as the mixed beds.

2.1.2 Type of Operation

All PWRs responding to the questionnaire used deep beds containing bead-type resin. In general, BWRs used powdered resins in the so-called "Powdex" type of operation for reactor coolant cleanup, although two of the nine respondents used deep bed operation.

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*P. L. Piciulo, "Technical Considerations for High Integrity Containers for the Disposal of Radioactive Ion Exchange Resin Waste," Draft Report, BNL-NUREG-30404, December 1981.

2.1.3 Temperature

The various demineralizer systems in the plants operate at reported temperatures of 75 to 150°F. Operating temperatures for the coolant cleanup beds are generally 100-120°F, with none reported over 140°F (60°C).

2.1.4 Criteria for Resin Replacement

Almost all the PWR's responding use too great a drop in decontamination factor (DF) as the principal criterion for replacing resin in the coolant cleanup bed. (DF is defined as the ratio of radioactivity in the influent to that in the effluent). The results of Clark's survey[2] are essentially the same. In that survey only one PWR (Maine Yankee) out of thirteen responding used something other than low DF as the main criterion. That exception was the use of "Li breakthrough or high radiation (100-300 R/h external to vessel)." It should be noted that in the present survey Maine Yankee reports using only low DF.

BWR's use mainly undesirably high pressure drop across the bed and undesirably high conductivities of the effluent. None reported using low DF as the main criterion. This is consistent with the results of Clark's survey where only one plant (LaCrosse) used low DF as its replacement criterion.

2.1.5 Relative Radiation Exposures and Evidence of Radiation Damage

Along with the request for information on criteria for resin replacement, we asked for the gamma-radiation field at the surface of resin bed vessels, and for any evidence of radiation damage to the resins. Values for the radiation fields were reported by several plants and are given in Table 2.1.

It is obvious that at the plants supplying these data, the primary coolant cleanup beds generate a radiation field many times greater than that of the beds in the other demineralizer systems. One would assume that this situation holds for all plants. This is expected since the primary coolant continuously flows through the high neutron flux of the reactor.

None of the plants responding to the survey reported any evidence of radiation damage to the resins. This is not surprising since spent resins are not stored for a long time after being taken out of service. In some plants, spent resins are shipped for disposal without storage, and the longest storage period seems to be about a year. Since the half-lives of the dominant radionuclides (RNs) with which the spent resins are loaded (Co-60 and Cs-137) are 5.3 and 30 years respectively, only a small fraction of the total dose eventually deposited will be received before shipment. It is interesting that one plant (Duane Arnold) reported relatively rapid gas pressure buildup in their drums of dewatered resins. This was due to bacterial action rather than radiation damage, since addition of formaldehyde, a biocide, could control the situation. To avoid the problem, they now solidify all their spent resins with cement and ship within a short period before any gas which might eventually form can build up appreciably.

Table 2.1

Radiation Fields at the Surface of Resin Bed Vessels

Plant	Field (R/h) at Bed in: Primary Coolant	Condensate or Boron Recycle	Radwaste	Fuel Pool
BWR's				
Dresden	50-200	1-3	1-5	10-100
Peach Bottom	---	---	0.3-0.6	7-12
PWR's				
Beaver Valley	$\leq$500	---	---	---
Donald C. Cook	500	0.5	---	35
Maine Yankee	500-1000	---	---	---
Yankee Rowe	5-100	---	---	$\leq$3
Zion	100	1	5	1

2.2 Activity Levels and Principal RNs in Liquid Streams

Questions were asked in the survey regarding the gross α- and β, γ-activities in the liquid streams before and after the various resin beds. It was also requested that the principal RNs in those streams be identified. A number of plants (8) did not respond to these questions, but most (15) gave data on gross β,γ-activity and some sent copies of analytical results giving a breakdown according to individual RN. A summary of the data on gross β, γ-activity in the different liquid streams is presented in Table 2.2. The principal RNs in the reactor coolant are listed in Table 2.3 for those reactors responding to that question. Amounts of the individual RNs are given in order of decreasing activity level in Tables 2.4-2.8 for the plants which sent detailed analysis sheets. The actual sheets are reproduced in Tables B-1 to B-4 of Appendix B. It should be noted that one of the analysis records sent (St. Lucie Plant, Unit 1) was a copy of Addendum II to Table 31 of Clark's 1978 report,[2] but the remainder were obtained during recent operations.

Table 2.2 indicates that activity levels in the various liquid streams do not vary widely from plant to plant, or from BWR to PWR. In general, the levels in the reactor coolant are considerably above those in the other streams, as would be expected. From Tables 2.4-2.8, the breakdown into individual RN activity levels shows that, in reactor coolant, the short-lived isotopes (mostly fission products and not necessarily in true solution) contribute essentially all the radioactivity, while the long-lived fission and activation products (Cs-137 and Co-60) generally contribute much less than 1%. Clark's survey[2] obtained detailed RN analyses from two BWR's and four PWR's, all

of them different from our responding plants except for Donald C. Cook and the aforementioned St. Lucie. The information obtained was very similar in extent and significance to that from the present survey.

In the present survey, as in Clark's, the detailed analyses show a considerable variation in the list of isotopes monitored at the different plants. Discussions with representatives of the plants indicate that there is, in fact, a wide variation in the proportions of the various RNs found in the coolant of different reactors. This variation is first of all due to the relative amounts of fission products compared to activation products, which in turn depend on a number of factors such as extent of fuel leaks, age of the reactor, and operating power level. Distribution of the more prominent RNs at a given reactor can change with time also, depending on reactor operating conditions, development of fuel leaks, and chemical treatment of the coolant. As an example of this last effect, addition to the coolant of hydrogen peroxide (which has been done at a number of plants) causes release of cobalt from the surface of reactor components in contact with the coolant. Thus, after such treatment, very large increases in the levels of Co-58 and Co-60 in the coolant will occur.

In addition to actual differences in RN distribution at different reactors, there is a difference in those RNs reported, because of specific analytical methods used. All plants appear to use γ-spectroscopy for analysis. Some plants record all RNs definitely identifiable. Others monitor only certain sets of RNs as determined by the computer software provided by the vendor of their analytical equipment. Their computer programs, after reporting amounts of the RNs being monitored, give the energies and count rates of all other γ-peaks observed. Thus, if any new RN shows up or an unusual increase is noted in any peak, appropriate additions can be made to the analytical procedure to keep track of the situation.

The results reported by Dresden and St. Lucie are the most extensive, and in terms of relative concentrations, are in reasonable agreement for those RNs common to both plants. The apparent discrepancy in the distibution of iodine isotopes (reversal of I-133 and -134 abundances, and the high I-131 level at St. Lucie) is probably indicative of release of the longer-lived isotopes (131 and 133) in a reactor trip at St. Lucie shortly before analysis. The other plants report mainly iodine and noble gas isotopes for their principal contributors to the gross β,γ-activity, and amounts of these isotopes are consistent with those reported by Dresden and St. Lucie.

A point to note regarding the relatively short-lived isotopes is that during reactor operation they will rapidly build up to steady state saturation levels on the resin beds and will thus constitute constant dose rate radiation sources, assuming constant reactor power. They could therefore deliver a significant radiation dose to the resins if the beds were kept in service for periods of several months. The magnitude of this effect is discussed in detail in a later section (3.5).

Table 2.2

Concentration of RNs in Feeds
to Different Demineralizer Resin Beds

Plant	Gross β,γ-Activity (μCi/mL) in: Primary Coolant	Condensate or Boron Recycle	Radwaste	Fuel Pool
BWR's				
Dresdsen	1		0.001	0.001
Edwin I. Hatch	0.1			0.001
Millstone	0.1-1	0.01		0.0001-0.001
Peach Bottom	0.05-0.2			0.0001-0.005
PWR's				
Beaver Valley	0.5	0.5	0.01	0.001
Donald C. Cook	0.19	0.19		0.004
Fort Calhoun	1.9			0.02
Kewaunee	0.1	0.01	0.005	0.01
St. Lucie	1.7[a]	0.06		0.002
Zion	0.05-0.5	0.005-0.5	0.001	

[a]From present survey. In Clark's 1978 survey, the value was 2.7, neglecting noble gas isotopes.

Table 2.3

Principal Radionuclides in Primary Coolant

Plant	Principal Radionuclides
BWR's	
Cooper	Co-58,60; Cr-51; Mn-54; Sr-92; Tc-99m; Ag-110m; Zn-65.
Dresden	Tc-101,104, I-132,133,134,135; Ba-139,141; Sr-91,92; Cs-137,138; Co-60.
Duane Arnold	Cr-51; Mn-54; Co-58,60; Fe-59; Na-24.
Millstone	Co-60; Cs-134,137; Fe-55; Mn-54.
PWR's	
Fort Calhoun	I-131,132,133,134,135; Cs-138; Co-58; Mn-54.
Donald C. Cook	Cs-138, Rb-88,89, Na-24, I-131,133; Y-88; Mn-54.
Kewaunee	Na-24; F-18.
Maine Yankee	Co-58,60; Mn-54; isotopes of iodine, cesium, and rubidium.
Point Beach	I-131,132,133,134,135; Cs-137,138; Rb-88; F-18; Na-24; Co-60.
St. Lucie	I-131,132,133,134,135; Rb-88; Mo-99; Cs-137,138; Te-132; Y-91; Co-60.
Yankee Rowe	Cs-138; I-131,132,133,134,135; Ba-139; Na-24; Mn-54; Se-75; Co-58; Nb-95.
Zion	Co-58,60; Cr-51; Cs-134,137,138; Ba-140; Mo-99; Zn-65; I-132.

Table 2.4

Concentrations of Radionuclides in Reactor Coolant at Dresden Unit 3 on July 21, 1981

Isotope	µCi/mL	Isotope	µCi/mL
Tc-101	4.6 E-1	Na-24	4.2 E-3
I -134	4.4 E-1	Cr-51	2.4 E-3
Tc-104	4.2 E-1	Co-60	1.3 E-3
Ba-141	1.8 E-1	Fe-59	5.9 E-4
Ba-139	1.7 E-1	Mn-54	4.4 E-4
I-135	1.4 E-1	As-76	3.4 E-4
I-132	1.3 E-1	Ce-141	2.9 E-4
Sr-92	8.6 E-2	Ru-103	2.7 E-4
Cs-138	6.9 E-2	Co-58	2.6 E-4
Tc-99m	6.5 E-2	Nb-95	1.6 E-4
I-133	5.7 E-2	Cs-136	1.5 E-4
Sr-91	3.2 E-2	Zr-95	1.5 E-4
Mo-99	9.5 E-3	Zn-65	7.3 E-5
Ba,La-140	6.3 E-3	Sb-124	6.5 E-5
Np-239	6.0 E-3	Cs-137	1.8 E-5
I-131	5.1 E-3	Ag-110m	7.7 E-6

Table 2.5

Concentrations of Radionuclides in Reactor Coolant at St. Lucie; Data Compiled for Clark's Report(2) in 1978.

Isotope	µCi/mL	Isotope	µCi/mL
Xe-133	2.49 E+1	Rb-89	8.80 E-3
Xe-135	1.64 E+1	Br-84	6.40 E-3
I-133	7.81 E-1	Co-58	6.40 E-3
I-132	5.48 E-1	Cr-51	5.20 E-3
I-135	3.72 E-1	Te-129	3.50 E-3
Kr-88	3.59 E-1	Cs-136	3.50 E-3
Rb-88	3.52 E-1	Te-134	3.06 E-3
Mo-99	2.80 E-1	Ba-140	8.43 E-4
Kr-85m	2.05 E-1	La-140	8.07 E-4
Xe-131m	2.04 E-1	Pr-143	8.06 E-4
I-132	1.50 E-1	Co-60	7.16 E-4
H-3	1.47 E-1	Sr-89	6.99 E-4
Kr-85	1.27 E-1	Ce-140	5.70 E-4
Kr-87	1.12 E-1	Ru-103	5.70 E-4
Cs-138	9.52 E-2	Sr-91	4.91 E-4
I-134	8.56 E-2	Y-90	1.41 E-4
Xe-138	4.97 E-2	Mn-54	3.79 E-5
Te-132	4.55 E-2	Sr-90	3.60 E-5
Cs-137	4.42 E-2	Ru-106	3.42 E-5
Y-91	1.53 E-2	Fe-59	2.94 E-5
Cs-134	1.38 E-2	Zr-95	1.29 E-6

Table 2.6

Concentrations of Radionuclides in Reactor Coolant at Donald C. Cook Unit 1.

Isotope	μCi/mL
Xe-133	1.56 E-1
Xe-135	6.49 E-2
Cs-138	3.60 E-2
Rb-88	1.53 E-2
Na-24	8.93 E-3
I-131	8.17 E-3
Kr-85m	8.01 E-3
Xe-133m	6.30 E-3
Kr-88	5.07 E-3
Kr-87	4.95 E-3
I-133	3.93 E-3
Y-88	3.82 E-3
Rb-89	3.35 E-3
Mn-54	1.98 E-3
Ar-41	1.89 E-3

Table 2.7

Concentrations of Radionuclides in Reactor Coolant at Fort Calhoun on August 17, 1981

Isotope	μCi/mL
Xe-133	9.70 E-1
Xe-135	1.96 E-1
Cs-138	1.28 E-1
I-133	8.07 E-2
I-132	7.52 E-2
I-134	6.15 E-2
I-131	5.12 E-2
Co-58	1.56 E-2
Mn-54	7.19 E-3
Ce-141	7.09 E-3
Nb-95	4.37 E-3

Table 2.8

Concentrations of Radionuclides in Reactor Coolant at Point Beach Unit 1 on January 20, 1981

Isotope	μCi/mL	Isotope	μCi/mL
I-134	4.54 E-1	I-131	3.10 E-2
Xe-135	3.29 E-1	Ar-41	8.83 E-3
Cs-138	2.78 E-1	Na-24	7.49 E-3
I-135	2.66 E-1	Kr-85	6.09 E-3
Rb-88	2.17 E-1	Cs-137	2.81 E-3
I-132	2.11 E-1	Mo-99	1.96 E-3
I-133	1.01 E-1	Cs-134	1.36 E-3
Kr-88	1.52 E-1	Te-132	8.74 E-4
Xe-138	1.37 E-1	Xe-133m	1.71 E-4
Xe-133	1.34 E-1	La-140	1.58 E-4
Xe-135m	9.26 E-2	Cs-136	1.16 E-4
Kr-85m	9.22 E-2	Co-58	9.58 E-5
F-18	6.94 E-2	Co-60	2.13 E-5
Kr-87	5.26 E-2	Mn-54	3.10 E-6
		Fe-59	3.04 E-6

2.3 Activity Loading and Principal RNs on Resins for Disposal

The surveys by Lin[1] and Clark[2] dealt mainly with performance of resins in treating liquid streams under nuclear power plant operating conditions. Our primary interest is in the effects of radiation both on resin performance and on properties of the resins after they leave the plants for final disposal. While we asked for activity levels in the liquid streams, as did Lin and Clark, we asked as well for the activity loadings on spent resins for disposal, which they did not. Thus any information they obtained on this subject was coincidental, and there is no section in their reports comparable to this one.

Table 2.9 summarizes information on the loadings of those resins with the highest activity levels in the plants listed. Table 2.10 lists the principal RNs on the resins as given by those plants answering the pertinent question. Tables 2.11-2.16 present analytical results from six plants which sent detailed analysis sheets giving a breakdown into individual RNs on the resins. These latter tables refer to reactor coolant cleanup resins, so it is likely that the data in Table 2.9 refer to similar resins. The actual analytical sheets are reproduced in Table C-1 to C-8 of Appendix C.

Perhaps the most striking thing about the data in Table 2.9 is the large variability between the different plants, both BWRs and PWRs. The loadings, both maximum and typical, show more than a hundredfold variation (maximum loading from 0.3 to 60 Ci/ft^3 and typical loadings from <0.1 to

30 Ci/ft^3). Also, the distribution of the relatively long-lived RNs varies from largely activation products (Co-60, Mn-54, etc.) to largely fission products (Cs-134,137) (see Tables A.2 and A.3).

Table 2.9

Typical and Maximum Activity Loading

Plant	Typical Loadings on Spent Coolant Cleanup Resin Ci/ft^3	Maximum Loading on Resin Shipped Ci/ft^3	Maximum Loading on a Bed Ci/ft^3
BWR's			
Dresden	4-5	18[a]	18
Duane Arnold	~0.6	0.15	1.55
Edwin I. Hatch	3-30	60[a]	60
Nine Mile Point	1	nr[b]	nr
Peach Bottom	0.1	maximum this year, 0.3	
Vermont Yankee	2-4	10	10
PWR's			
Beaver Valley	5	10	14.8
Donald C. Cook	nr	6.4	17
Joseph M. Farley	<0.1	3	7
Maine Yankee	~1	125[c]	nr
Point Beach	0.1-1	33	slightly >33
Prairie Island	nr	1	2
St. Lucie	nr	6	6
Trojan	~2	5.4	5.4
Yankee Rowe	nr	3	20
Zion	1-5	30	40

[a]Before solidification.
[b]nr - no response.
[c]Mostly Co-58 (71-day half-life).

Table 2.10

Principal Radionuclides on Resins[a]

Plant	Principal Radionuclides
BWRs	
Dresden	Co-58,60; Mn-54; Cs-134,137.
Edwin I. Hatch	Cs-134,137; Zn-65; Co-60.
Peach Bottom	Zn-65; Co-58,60; Cs-134,137; Cr-51; I-131; Mn-54.
Vermont Yankee	Cs-134,137; Zn-65; Co-58,60; Zr-95; Mn-54.
PWRs	
Donald C. Cook	Co-58,60; Cs-134,137; Mn-54; Sb-124.
Joseph M. Farley	Co-58,60; Mn-54; Cs-137; H-3.
Maine Yankee	Co-58,60; Mn-54; Cs-134,137; Sr-89,90.
Point Beach	Co-58,60; Cs-134,137; Sb-125; Ru-106; Mn-54; Cr-51.
Trojan	Co-58,60; Cs-134,137; Mn-54; Ce-144; Zn-65; Nb-95; Sb-125; Sr-90.

[a] Usually coolant cleanup resin, but not always specified.

Table 2.11

Levels of Radionuclides on Resin for Shipment at Donald C. Cook on June 21, 1981

Isotope	μCi/mL[a]
Co-58	1.55 E-1
Co-60	5.48 E-2
Cs-137	1.44 E-2
Sb-124	8.88 E-3
Cs-134	8.87 E-3
Mn-54	8.03 E-3
I-131	3.43 E-3
Nb-95	1.04 E-3
Co-57	7.03 E-4

[a] 1 Ci/ft^3 = 35.3 μCi/mL.

Some fission and activation products can be present at relatively high percentages at one plant, but not at others. For example, the activation product Zn-65 is the predominant activation product at two plants, in one of which (E. I. Hatch, Table 2.12) fission products constitute the bulk of the activity, and in the other (Peach Bottom, Table 2.13) activation products predominate. In most other plants (Tables A-1 and A-2) Zn-65 is not mentioned. Although Co-58 is normally present in smaller amounts than Co-60, it exceeds Co-60 at Donald C. Cook (Table 2.11) by a factor of 3, and at Maine Yankee (Table 2.13) by a factor of nearly 20. In the case of fission products, Ce-144 is an example of one that is present in an amount comparable to Cs-134 and Cs-137 at Trojan (Table 2.15) but is not significant at most other plants.

Table 2.12

Levels of Radionuclides on Unit 1
Coolant Cleanup Resin at E. I. Hatch.

Isotope	μCi/g
Cs-137	7.57 E+2
Cs-134	5.46 E+2
Zn-65	1.28 E+2
Co-60	3.01 E+1

Table 2.13

Levels of Radionuclides on Resin for
Shipment at Maine Yankee on March 11, 1980

Isotope	μCi/g
Co-58	1.70 E+2
Cs-137	2.11 E+1
Cs-134	1.11 E+1
Co-60	9.76 E 0

Table 2.14

Levels of Radionuclides on Resin
for Shipment at Peach Bottom

Isotope	μCi/g
From Analysis	
Zn-65	2.45 E 0
Co-60	6.64 E-1
Cs-137	4.06 E-1
Cs-134	2.86 E-1
Cr-51	2.06 E-1
I-131	1.27 E-1
Co-58	1.05 E-1
La-140	2.60 E-2
Mn-54	1.10 E-2
Estimated (pure β-emitters)	
Sr-90	4.06 E-3
Fe-55	7.64 E-1
Ni-63	5.98 E-1

Table 2.15

Levels of Radionuclides on Resin
From Trojan on May 14, 1981

Isotope	μCi/g
Co-60	4.57 E+1
Cs-137	1.26 E+1
Cs-134	5.13 E 0
Mn-54	2.78 E 0
Co-58	5.32 E-1
Ce-144	3.49 E-1
Ru-106	1.95 E-1
Sb-125	1.65 E-1
Cr-51	1.21 E-1
Zn-65	6.71 E-2
Co-57	6.07 E-2
Fe-59	5.87 E-2
Ag-110m	4.04 E-2
Nb-95	2.26 E-2
Sb-124	1.78 E-2
Ce-141	1.26 E-2

Table 2.16

Levels of Radionuclides on Reactor Cleanup Resin at Vermont Yankee

Isotope	μCi/g
Cs-137	4.84 E+1
Co-60	3.05 E+1
Zn-65	2.03 E+1
Cs-134	1.78 E+1
Zr-95	1.11 E+1
Mn-54	5.98 E 0
Co-58	5.88 E 0
I-131	5.35 E-1
Tc-99m	4.19 E-1

Variations of this nature are expected due to different operating conditions at different plants, and the long term radiation doses delivered to the resins by all of these other RNs is small (due to their shorter half-lives) relative to the dose from Co-60 and Cs-137. What is perhaps unexpected is the absolute magnitude of some of the maximum loadings reported, higher than 30 Ci/ft^3 at 3 of the 16 plants. A fourth plant, Maine Yankee, reported 125 Ci/ft^3 for the maximum loading on a resin for disposal, but this turned out to be due largely to Co-58* which has a half-life of only 71 days and would thus require much higher loadings to cause significant radiation damage.

E. I. Hatch reported the highest loading of radioactivity with a half-life >5 years. That loading (60 Ci/ft^3) was composed largely of Cs-137 and was therefore comparable to that encountered on a resin prefilter used in the AFHB cleanup at TMI-II.[8] The latter was loaded to approximately 40 Ci/ft^3, assuming uniform loading, and was considered to be loaded to a higher than desirable level in terms of then-known power plant practice, and from the point of view of radiation damage to the resin. The maximum loading on waste shipped from E. I. Hatch would have been <60 Ci/ft^3 because of dilution when the resin was solidified. They ship only 100 ft^3/yr of coolant cleanup resin for each reactor, but 15,000 ft^3/year, for each reactor, of their other resins, which are loaded to only 0.015-0.25 Ci/ft^3. Thus this plant could use many more times the amount of coolant cleanup resin than they presently use (thereby considerably reducing their loadings) without appreciably increasing their total resin use. Another acceptable way of reducing long-term dose to their resins might be to mix the coolant cleanup resins with some of the other resins before solidifying.

*D. R. MacKenzie and K. J. Swyler, Memo to File, March 1, 1982, "Report of Trip to Maine Yankee Atomic Power Plant, February 23, 1982."

The other two plants reporting loadings >30 Ci/ft^3 are Zion and Point Beach, with 40 and approximately 33 Ci/ft^3 respectively for maximum loading on a bed. Since Zion solidifies for shipment, their maximum loading on shipped waste would have been less than the reported 30 Ci/ft^3 reported as maximum. All the other plants report 10 or fewer Ci/ft^3 as the maximum loading on a resin shipped to date. This includes Dresden, since their value of 18 is for the resin before dilution by solidification. Duane Arnold reported a maximum of 0.15 Ci/ft^3 with an annual shipment of 6000 ft^3 and Peach Bottom 0.3 Ci/ft^3. The others (9 out of 15) were all in the 1-10 Ci/ft^3 range.

The maximum values reported were often termed unusually high for the particular plant, or were said to refer to a one-time shipment. Indeed, 1-10 Ci/ft^3 is more typical of the loadings encountered in practice, e.g., 7 out of the 12 plants for which information is available gave figures in this range (Table 2.9). Four plants have typical loadings that are even lower, and even E. I. Hatch, the only plant reporting >10 Ci/ft^3 for normal shipment gives a range of 3-30 Ci/ft^3. It can be concluded, then, that U.S. commercial reactors typically ship coolant cleanup resin in the range of 1-10 Ci/ft^3 or less, but occasionally a few may significantly exceed 10 Ci/ft^3.

2.4 Sr-90 in Power Plant Wastes

For routine analysis most power plants determine the β,γ-activity in liquid and resin samples by γ-counting, often with energy analysis to identify and measure the individual RNs (see, for example, Tables 2.11 - 2.15). This counting technique is very convenient, and particularly suited to routine analysis. However, it completely misses the pure β-emitter Sr-90 (29 yr half-life) and its short-lived daughter (2.7 day half-life) Y-90, an almost pure β-emitter. Because of its high β-energy (2.28 MeV maximum energy), Y-90 is responsible for most of the radiation damage, toxicity, etc. from the Sr,Y-90 couple when they are in secular equilibrium. Curie for curie, Sr,Y-90 delivers approximately twice the long-term dose delivered by Cs-137. It is thus essential to know the Sr-90 loading on a resin in order accurately to determine long-term dose.

In the responses to our questionnaire, three plants mentioned Sr-90 - Trojan, Peach Bottom and Maine Yankee. Trojan and Maine Yankee list it as one of the principal RNs on spent resin (see Table 2.10). Trojan submitted an analysis done in July 1981 by Science Applications, Inc., which found 0.32 μCi Sr-90/g in a resin sample containing 12.6 μCi Cs-137/g. The Sr-90 content thus amounted to 2.5% of the Cs-137 in the sample. Analytical results from Peach Bottom gave an estimated value for Sr-90 exactly 1% of that for Cs-137. When asked about this, personnel at Peach Bottom stated that they analyzed samples periodically for Sr-90, and the amount has never exceeded 1% of the Cs-137 present. They, therefore, use that value routinely as a conservative estimate which they are confident will not be exceeded. Possible requirements, as a result of this kind of experience, for dealing with a maximum loading regulation, are discussed in Section 3.4.3.

2.5 TRU Isotopes on Spent Resins

The questionnaire requested information on α-activity in the liquid streams and on spent resin. Most respondents either did not answer these questions (11) or indicated that any α-activity was below minimum detectable levels (8). Four plants had measurable amounts of α-activity associated with resins, Kewaunee, Millstone, Maine Yankee and Trojan.

Kewaunee and Millstone reported concentrations in the liquid streams to be less than the minimum detectable, and noted that the α-activity on their resins was <10 nCi/g. Maine Yankee gave <1 μCi/mL for the α-activity associated with resins. Since 1 μCi/mL is many times higher than the 10 nCi/g TRU limit, personnel at Maine Yankee were questioned about their response. They stated that our questionnaire asked for μCi/mL, and they gave the value "<1" to indicate that their activity was not in the μCi/mL range. In fact, it was barely detectable and levels were well below 10 nCi/g on all resins shipped for disposal.

Trojan sent a copy of a typical recent analysis, which is reproduced in Appendix D as Table D-1. The data are presented in Table 2.17 in terms of nCi/g. Since the concentration of TRU isotopes on the resin sample was approximately 14 nCi/g, the resin would be technically TRU waste, being above the 10 nCi/g TRU limit. Trojan disposes of its spent resins by solidification, so that for this particular batch, its α-activity after solidification was 4.6 nCi/g of solid waste, well below the TRU limit. So far all their resin wastes have had no trouble meeting the requirements of non-TRU waste.

Table 2.17

Alpha Emitting Nuclide Levels on a Filter and a Resin Sample at Trojan

Nuclides	Amount on filter nCi Total	Concentration on Resin nCi/g
Pu-239,240	1.31	11.4
Pu-238	0.58	3.0
Am-241	0.015	0.060
Cm-242,243	0.051	0.073
Cm-244	0.004	0.006

On the basis of the responses to the survey, it can be concluded that levels of TRU isotopes on spent resins from commercial reactors are very low, either below 10 nCi/g of resin, or of such a value that the level in the final waste form can easily be kept below 10 nCi/g so that the waste does not constitute TRU waste. Such levels of α-activity produce radiation doses to the resin that cause insignificant amounts of damage.

2.6 Disposal Information

2.6.1 Methods

The plants responding to the questionaire used a wide range of disposal methods, from shipping dewatered resin in containers of approximately 6 ft^3 (55-gal drums) to 200 ft^3 (steel liners), to solidifying in 55-gal drums and larger steel container. Several plants contracted out all their resin disposal to commercial firms, which normally solidify, so presumably the resins from these plants are being solidified. A fair proportion indicated they planned to use HIC's for disposing of dewatered resin. The plants which did their own solidification generally used cement.

The solidification option is useful for plants where activity levels are high (either β, γ, as considered in Section 2.3, or α, as considered in Section 2.5). The dilution occurring during solidification makes the resultant waste less of an exposure risk for workers if β,γ-activity is involved, or can keep the final product from being classed as TRU waste if α-activity is the limiting factor. This idea should not be carried too far, but the published information indicates that composites of resin with cement, for example, do not suffer any deleterious affects from irradiation to doses of between 10^8 and 10^9 rad.(5) Doses of 10^8 rad to a composite containing 50% resin by volume would require a Cs-137 loading in the range of 30-40 Ci/ft^3 on the original resin, depending on the size and shape of the composite waste form.

2.6.2 Amounts

There is a very wide variation in amounts of spent resin shipped annually per reactor. The values reported in the survey are given in Tables A.1 and A.2. BWRs in general report shipping ten or more times as much as PWRs. Amounts from BWRs ranged from 1,000 to 25,000 ft^3, except for Dresden which reported only 300 ft^3. For PWRs, amounts were mostly 100 to 300 ft^3, with only Zion reporting an amount (5,000 to 10,000 ft^3) typical of a BWR. Of those shipping waste with radionuclide loadings of 10 Ci/ft^3 or greater, several plants had low total shipments, including Dresden, the only BWR reporting a low amount shipped. The remaining three plants shipped large amounts. It has already been mentioned (Section 2.3) that Edwin I. Hatch reported shipping 100 ft^3/yr of reactor coolant cleanup resin out of a total annual shipment of 15,000 ft^3. Two other plants, Zion (PWR) and Vermont Yankee (BWR) reported shipping several thousand ft^3 annually. They did not give a breakdown to indicate the amount of coolant cleanup resin shipped.

3. SELECTION OF A MAXIMUM PERMISSIBLE LOADING

NRC in a recent draft branch technical position (BTP) for guidance to waste generators proposed setting a maximum of 10 Ci/ft^3 for the RN loading on organic ion exchange resins being shipped for disposal. At this loading of Cs-137, a long-term dose of approximately 10^8 rad would be delivered to the resin. In this section, the rationale for choosing 10 Ci/ft^3 maximum loading is examined, and it is concluded that it is desirable for relatively long-lived RNs such as Cs-137 and Sr-90. However, because of the variation in disintegration energy and half-life (and therefore in total dose delivered) of the RNs found on waste resins in practice, the 10^8 rad total dose limit is considered a more appropriate basis for a guideline.

3.1 Need for a Loading Limit

Ion exchange resins suffer degradation on exposure to sufficiently high doses of radiation. Such degradation results from damage to the polymer structural framework and removal of functional groups by radiation decomposition reactions. The former causes formation of hydrogen and other gases. The latter can cause not only gas formation, but loss of exchange capacity, and changes in pH which might enhance corrosion of metal containers. Also radiolytic decomposition of the water remaining in dewatered resins produces gas, as well as reactive radicals capable of attacking the polymer structure and functional groups. These effects should obviously be avoided in wastes for consignment to a burial site, hence the need for an upper loading limit below which their magnitude will be insignificant.

It must be realized that effects due to loading with the RNs commonly encountered at power plants will take a relatively long time to show up at realistic loading levels, i.e., at levels low enough to permit normal handling and not lead to problems with worker exposure during handling. The spent resins are not stored for long periods (maximum about a year, and sometimes not at all) at nuclear power plants. Therefore, the total absorbed dose will not be large, and possible damage will not yet be apparent, before the resins are shipped for disposal. It is only after a period of many years that total dose to the resins will reach the stage where radiation damage would be obvious, in other words, long after the resins have been deposited at the burial site. In the case of Co-60, for example, in one year only 12% of the eventual total dose will be delivered, while for Cs-137 the amount will be only 2.4%, and even for Co-60 it will take over 5 years to reach 50% of the eventual total dose.

3.2 Literature Information on Radiation Damage

On the basis of information in the literature survey of Gangwer, Goldstein and Pillay,[4] polymerization-type resins apparently are superior to condensation-type resins in terms of chemical, mechanical and thermal properties, although their radiation stability is not as great. In discussions with nuclear power plant personnel during preparation of our questionnaire, the point was made that utilities choose resins for plant operations that will

provide them with maximum use, or service time. That means presumably, that their choice would normally be polymerization-type with its better chemical, mechanical, and thermal properties. Radiation stability is not an important factor during service so it will not be the criterion on which the choice is based.

The results of the survey confirm this, and we conclude that the resins used throughout the nuclear power industry are the strong acid cation exchangers (with sulfonic acid functional groups) and the strong base anion exchangers (with quaternary ammonium ion functional groups). Extent of cross-linking of the polystyrene with divinylbenzene (DVB) in cation exchangers varies from 4-16% in commercially available resins, and the power plants appear to use mainly 8 or 10% cross-linked material. For purposes of this report then, these particular resins are the ones for which information on radiation damage is most desired, and the ones which must be considered the most important in determining a maximum loading requirement.

3.2.1 General Principles

Exact mechanisms to explain the interaction of radiation with different types of organic ion exchange resins have not been determined, due in large part to the great complexity of the polymer systems.(4) However, in a qualitative way, the kinds of chemical bonds attacked and the relative degrees to which different types of chemical reactions occur are known. Also, physical and chemical changes as a result of irradiation of the resins are reasonably well known in terms of changes in resin properties.

The principal types of bonds attacked in the resin molecules are: the C-C bonds in the polymer structural framework, the bonds linking the functional groups to the carbon framework, and the C-H bonds. The bonds linking functional groups to carbon atoms are the most susceptible to breakage. Scission of these bonds leads to loss of exchange capacity. Scission of C-C bonds leads to degradation of the polymer chains and to breakage of cross-links, thereby partially breaking down the three-dimensional structural network. All these processes are destructive of the resin molecules, leading to undesirable changes in properties. Scission of C-H bonds leads to formation of hydrogen gas (not destructive in itself) and to a certain amount of cross linking (constructive rather than destructive). This latter process therefore mitigates to some extent the damage done by other processes, but the overall effect of radiation is deterioration of the resin. This is particularly true in the presence of water and air, when oxidative processes also occur.

Some of the properties and the differences in susceptibility of the different types of resins are noted below. These are, however, broad generalizations, and there will undoubtedly be exceptions.

1. Most cation exchangers begin to show significant damage at a dose of around 10^8 rad, while most anion exchangers are damaged noticeably at a somewhat lower dose. The one type of resin with a

much greater radiation resistance (an anion exchanger with pyridine groups in the structural framework) is not used by power plants as far as is known.

2. The effects of radiation which are of most concern to disposal of spent resins are: gas generation, changes in pH, agglomeration, enhancement of corrosion of metals in contact with the resins, and, to a lesser extent, loss of exchange capacity.

3. Larger changes in properties (up to a factor of five greater) are observed when resins are irradiated in the presence of water than when they are irradiated dry. The stabilities of the polystyrene-divinylbenzene resins irradiated in the presence of water show the following order:

   pyridine based anion exchanger > nuclear sulfonic and carboxylic cation exchanger > weak base anion exchangers of the primary, secondary, and tertiary type > strong base anion exchanger of the quaternary ammonium type.

4. In general, the salt forms of both cation and anion exchangers are more resistant to radiation than are the H^+ and OH^- forms.

5. There appears to be no difference in irradiation effects between the different types of low LET* radiation sources (X-rays, γ-rays, fast electrons, and β-particles) as regards either differences in the nature of the radiation (wave vs particle) or whether the irradiation is internal or external (β-particle vs the remainder).

3.2.2 Effect on Exchange Capacity

The exchange capacity of a resin is a measure of the extent to which it can remove ions from solution, and is expressed as milliequivalents per gram of resin. For the strong acid cation exchanger of most interest to this report, absorption of radiation causes cleavage of sulfonic acid groups. The result is formation of SO_2 and consequent loss of the exchange capacity which the SO_3H^- groups had provided. In the presence of oxidants, some carboxylic acid and phenolic OH groups will be formed, and at low doses total exchange capacity (TEC) due to these groups plus the sulfonic acid groups may increase slightly. However, at higher doses, the net result is overall loss of TEC.

Although the qualitative picture seems clear, experimental results have not generally yielded quantitative interpretation of the data so as to enable

*LET, or linear energy transfer, refers to the density of energy deposition along a particle path.

differences between the results of different studies to be explained.[4] For example, several workers found that radiation resistance, as measured by loss of TEC, increased with increasing degree of cross-linkage of the starting material. However, Tulupov et al.[10] reported no significant change in radiation stability of the resin KU-2--again measured by TEC--going from 8% to 24% cross-linkage.

Results of a study[11] of the effect of cross-linkage of Dowex 50 on loss of sulfonic acid groups are summarized in Table 3.1. This shows the variation in G-value for loss of SO_3H^- groups with dose for four degrees of cross-linkage from 4% to 16%. (G-value is defined as the number of radicals or molecules formed or decomposed per 100 eV absorbed.) The resin stability increases (i.e., G-value for loss of SO_3H^- decreases) from 4% cross-linkage to 12% and then decreases at 16%, except for the highest dose of nearly 10^9 rad. For any degree of cross-linkage, the G-value decreases with dose.

Table 3.1

G-Value for Loss of Sulfonic Acid Groups as a Function of Dose for Dowex 50 of Various Cross-Linkages[12]

	G-value, Number of SO_3H^- Groups/100 eV			
Dose (rad)	Dowex 50 x 4	Dowex 50 x 8	Dowex 50 x 12	Dowex 50 x 16
2.2×10^8	3.2	2.0	1.5	2.4
4.7×10^8	1.7	1.3	1.3	1.5
8.6×10^8	1.4	1.1	1.0	1.0

The point that is of concern to this report, however, is the high G-values observed for all the cross-linkages at the lowest dose of 2×10^8 rad. Another, perhaps simpler, way of expressing the extent of this radiation damage is to present it in terms of percent loss of exchange capacity. Results of several studies using Dowex 50, Amberlite IR-100, and Dowex 50W showed that at a dose of 10^8 rad approximately 5-10% of the exchange capacity was lost.[12-14]

Anion exchangers also lose capacity on irradiation. An effect somewhat analogous to that occurring with cation resins is observed with strong base anion exchangers, in that at low doses TEC is lost only slowly because some of the quaternary ammonium groups attacked are converted to weakly basic amino groups. However, with increasing dose net TEC decreases. As with cation exchangers, presence of water and salts exerts a fairly large effect, but again the spread in experimental results is too great to permit satisfactory interpretation of the observed effects.

G-values for loss of anion exchange capacity are found largely in the Russian literature.[4] They are comparable to, but generally higher than, those for the strong acid cation resins. In terms of percentage loss of exchange capacity on irradiation, Kazanjian et al.[15] with the strong base anion resin Dowex-1 found decreases in capacity of well over 10% at 10^8 rad. In fact, the decrease normally was considerably greater than that, and about double that observed for Dowex 50 strong acid cation exchanger. Thus, overall, it must be concluded that both anion and cation exchangers are quite susceptible to radiation damage as indicated by loss of exchange capacity.

3.2.3 Gas Generation

Somewhat less work has been reported on gas generation during resin irradiation than on the loss of exchange capacity. Most of the results reported are for cation exchangers.[4] In particular, Mohorcic et al.[16,17] and Kazanjian and Horrell[18] have made quantitative determinations of the different gaseous products formed during radiolysis. Of the few published results for anion exchange resins, those of Kazanjian[15] give quantitative estimates of the gaseous products.

The principal gases formed during irradiation of resins are hydrogen and carbon dioxide. Other gases reported to be formed from cation exchange resins are CO, SO_2, O_2, and CH_4, and from anion exchangers, CO, N_2, NH_3, N_2O, and NO. G-values for formation of several of the gases are given in Table 3.2. The G-values are noticeably much smaller than those for loss of exchange capacity, except for samples immersed in water or 7 M HNO_3. For cation exchangers in water, $G(H_2)$ is between 1 and 2 and $G(CO_2)$ is 0.4. For anion exchange resin in 7 M HNO_3, $G(CO_2)$ is 0.6. In general, G-values for formation of hydrogen are 0.1 or less, and those for the other gases considerably lower. There is a very wide variation in $G(SO_2)$, which is not readily explainable by an obvious factor such as solubility of SO_2 in the water of the resin matrix. Yields of the other gases normally are smaller when the resins are irradiated dry. Irradiation of cation resin in the salt form (Na^+ and Li^+) did not seem to make an appreciable difference, except for the Li^+ form in the dry state, where the G-values obtained for all gases except SO_2 were lower than under any other conditions.

The amounts of H_2, CO_2, and CO formed by radiolysis are all approximately linear with dose to total doses $>10^8$ rad. This is shown in Figure 3.1 for H_2 production from 10% cross-linked Dowex 50W cation resin.[16] The data for this figure are taken directly from Reference 16 since several of the plots in Figure 11 of Gangwer, Goldstein, and Pillay[4] are incorrectly reproduced from the original. The Li^+ form was found to be somewhat less stable than the H^+ form when swollen with water, but slightly more stable when immersed in water. The data for immersion in water cannot be conveniently plotted on the same graph because the yields are so much higher.

There is an extremely large difference reported in H_2 yield (factor of 100 - Table 3.2 and Figure 3.1) between the Li^+ form dry and swollen. This difference is difficult to understand, for one thing because the difference between the H^+ form dry and swollen is roughly a factor of 3. Such a

magnitude is not unexpected in view of the additional H_2 production from radiolysis of the water in the matrix of the swollen resin. This published report of extreme suppression of H_2 production in the dry Li^+ form thus appears to be suspect, especially since Li^+ does not suppress H_2 production in the moist resin, and the yields of the other gases are not greatly different between the Li^+ form dry and swollen. Aside from this one anomalous result, the data reported seem quite acceptable.

Table 3.2

G-Values for Gaseous Product Formation During the Radiation Decomposition of Ion Exchange Resins[a]

Resin	Ionic Form	Irradiation Condition	$G(H_2)$	$G(SO_2)$[b]	$G(CO_2)$	$G(CO)$	Ref. No.
Dowex 50W x 4	H^+, Na^+	Air-dry and 0.1 N (HCl)	0.1	--	--	--	19
Dowex 50W x 10	H^+	Dry	0.026	0.087	0.035	0.009	17
Zeo-Karb 215[b]	H^+	Dry	0.051	0.001	0.019	--	17
Dowex 50W x 10	H^+	12-47% water	0.04 to 0.12	~0.14	~0.023	~0.022	18
Dowex 50W x 10	Li^+	Dry	0.001	0.26	0.07	0.002	17
Dowex 50W x 10	Li^+	Swollen	0.11	0.5	0.008	0.020	17
Dowex 50W x 10	H^+	Swollen	0.095	0.006	0.019	0.027	17
Zeo-Karb 215[c]	H^+	Swollen	0.12	0.005	0.046	0.005	17
Zeo-Karb 215[c]	H^+	Immersed in water	1.7	0.002	0.43	--	17
Dowex 50W x 10	H^+	Immersed in water	1.7	--	0.41	--	17
Dowex 50W x 10	Li^+	Immersed in water	1.3	--	0.41	--	17
Dowex 1 x 4	NO_3^-	Air-dry	0.09	--	--	0.002	16
Powex 1 x 4	NO_3^-	In 7N HNO_3	0.02	--	0.6	--	16

[a]Taken from Table VII of Gangwer, Goldstein and Pillay.(4) Note that in Gangwer et al., the data of line 6 have been incorrectly reproduced from the original.
[b]G-values for the lowest absorbed dose.
[c]Condensation-type resin.

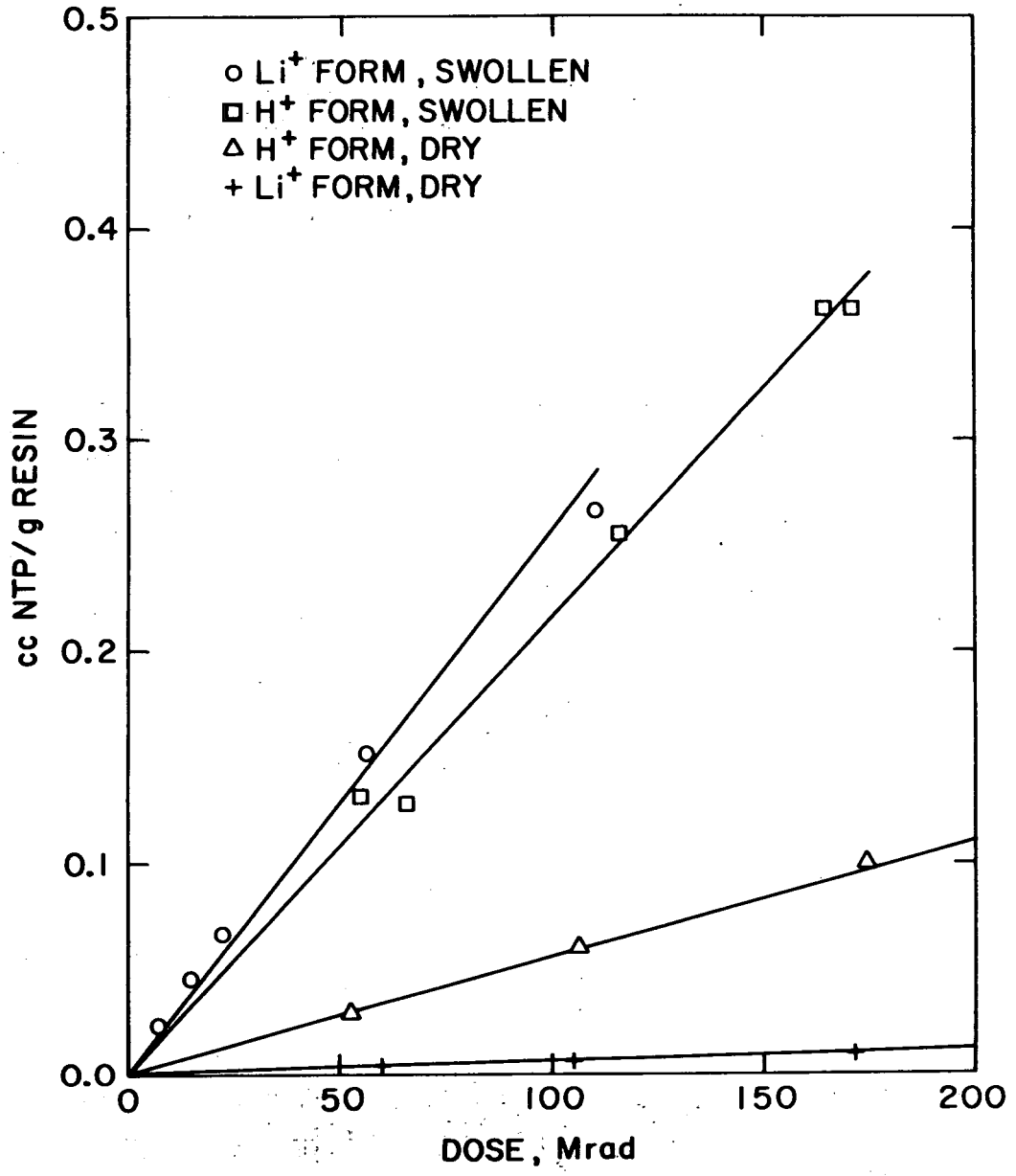

Figure 3.1 Hydrogen production from irradiation of 10% cross-linked Dowex 50W. Results reported by Mohorcic and Kramer,[16] 1968.

3.2.4 Work Related to TMI-II Cleanup

Recent work[5-8] in connection with the TMI-II cleanup program has been mentioned in the introduction. This work has provided useful information on the effects of radiation to total doses $>10^9$ rad on various properties of the resin types used by the nuclear power industry. The effect on exchange capacity has not been investigated, but gas generation, change in pH, and several other chemical and physical properties have been studied.

3.2.4.1 Gas Generation

In discussion of gas generation during irradiation, it should be pointed out that, while there is a net formation of gaseous products, there is a considerable decrease in the amount of any oxygen present. According to work at BNL[5] on effects of irradiation on the resins used in the cleanup of TMI-II contaminated water, this oxygen depletion is quite rapid in the earlier stages. Thus in the experimental air irradiations of the proprietary formulation given the name D-mix, both by itself and incorporated in a solidified form with cement, an intitial pressure decrease was observed. Although the gas over the sample was not analyzed during this period, almost no oxygen was left at the end of the irradiations, and oxygen consumption presumably was responsible for the initial pressure drop. This effect was not observed in the earlier work reviewed by Gangwer et al.,[4] and of course it would not occur in irradiations in vacuum or under an inert gas cover. However, generation of oxygen is not repored for cation exchange resins by most workers, and not at all for anion exchange resins, and this lack of oxygen production is compatible with the oxygen depletion observed in BNL's irradiation in the presence of air.

In the TMI-II related work, gas generation was studied at BNL[5,8] and by McFarland.[6,19] Hydrogen and methane were observed by BNL[8] on irradiation of the nuclear grade cation exchanger IRN-77. No special study of individual gas yields was carried out. In the experiments described in the last paragraph in which oxygen depletion was observed, BNL measured the amounts of H_2, CH_4 and CO_2 formed.[5] The values obtained were useful for comparative purposes only, since the resin irradiated was a proprietary formulation. McFarland irradiated the cation exchanger Dow HCR-S, Na^+ form, and the anion exchanger Dow SBR-OH, BO_3^{-3} form, to high doses in an experiment where buildup of gas pressure was followed.[19] Pressures were incorrectly converted to moles (or volumes) because of inaccurate estimation of void space in the experimental systems. Subsequent work[6] enabled determination of the void space and correction of the volumes produced at STP.

The pressure plots are reported in Figures 7 and 8 of Reference 6. The data for both resins below 1.6×10^9 rad are plotted on one diagram in Figure 3.2. Although these yield-dose curves of McFarland are nearly linear, as were those of earlier workers, there are definite increases in the slopes as one goes to higher dose. The data show that the anion exchanger exhibits a rate of gas production several times that of the cation exchanger (about 8 times when the larger void volume in the anion capsules is taken into

account). Both plots indicate a threshold for gas production somewhere around 5×10^7 rad. This is contrary to the results in the literature reviewed by Gangwer, Goldstein, and Pillay,[4] and, in fact, McFarland's plots of yield-dose curves for hydrogen and methane do not show a threshold. The threshold for pressure buildup is most likely due to the oxygen depletion effect observed by BNL, and it is significant in this regard that pressures in the BNL experiments, after their initial decrease, returned to their original values and began to increase at a dose in the vicinity of 5×10^7 rad.

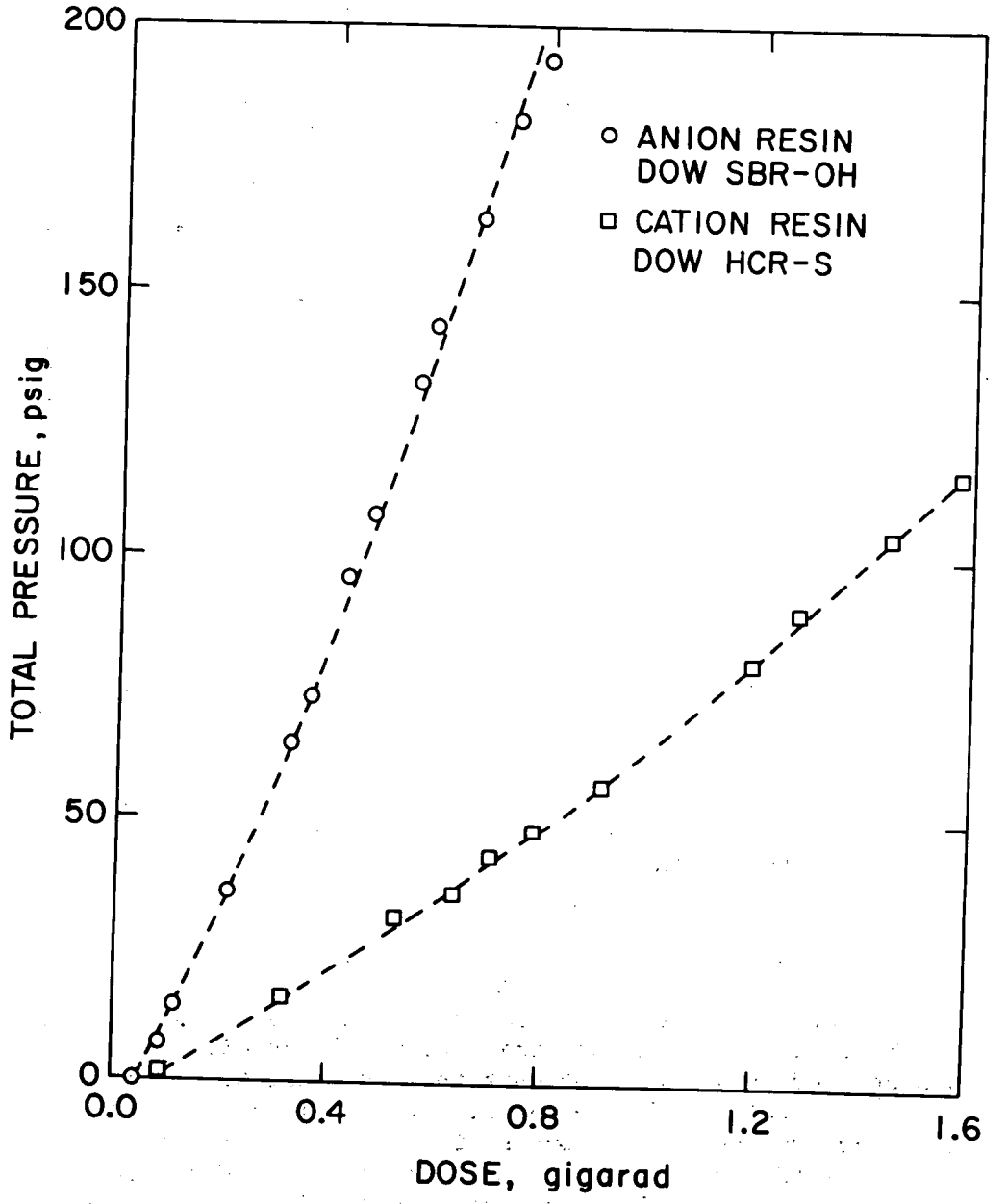

Figure 3.2 Gas pressures developed on irradiation of cation and anion resins. Results reported by McFarland,[6] 1981.

The gas in McFarland's experiments was analyzed at the end of the irradiations. In Table 3.3, results are given for the anion exchanger as reported in Table 5 of Reference 6. The results given for the cation exchanger in Table 3.3 were taken from the same source, but adjusted downward to the same dose (7.9 x 10^8 rad) as the anion resin had received. For the adjustment, the actual experimental curve of pressure vs dose given in Figure 3.2 was used. The distribution of gases for the two resins is similar, but the overall amounts are much greater for the anion resin--almost a factor of eight for total gas.

Table 3.3

Yields of Product Gases[a] From 7.9 x 10^8 rad Irradiation of Dewatered Cation and Anion Exchange Resin
Gas Volume, cc/g Resin

Resin	Total Gas	H_2	CO_2	CO	CH_4	O_2	N_2	Sulfur Gases µg S/g Resin	G_{total}
Cation - Na^+ form of Dow HCR-S	1.7	0.70	0.21	0.25	0.03	0.18	—	0.02	0.09
Anion - BO_3^{-3} form of Dow SBR-OH	12.6	6.8	1.1	1.3	0.81	0.56	0.68	—	0.68

[a]Data taken from Table 5 of Reference 7. Yields for cation exchanger estimated from those at 2.5x10^9 rad using actual yield-dose plot of Figure 3.2, and assuming the same proportion of individual gases.

The G-values for total gas are also given in Table 3.3. They agree reasonably well with the results of work reported in the last section (3.2.3). The G-value for H_2 production for the anion exchanger at 10^8 rad (0.15) is about double that of Kazanjian's[15] for dry Dowex-1, which is a reasonable difference between moist and dry resins. The result for the cation exchanger is close to that for Mohorcic's[16] dry Dowex 50W in the H^+ form, and much lower than that for the swollen Dowex 50W in the H^+ form. This could be due to McFarland's resin being in the Na^+ form and/or differences in the amount of water present in the resins in the two studies.

3.2.4.2 Changes in Appearance

Several effects have been observed, such as swelling and agglomeration, which are probably partly physical and partly chemical in nature. Swelling was studied by a number of workers, and their work reviewed by Gangwer et al.[4] Swelling and shrinking effects are best observed in irradiation of resins immersed in aqueous media--often swelling occurs up to moderately high doses (3-4 x 10^8 rad), and at still higher doses the resin will shrink. The effect would probably not be of significance for dewatered resins at doses somewhat higher than 10^8 rad, since there would be no source of water for the resins to take up. However, at higher doses, some of the water

in the resins would presumably be released because of shrinkage, and could be a problem for metal containers if it were corrosive.

Agglomeration has been examined in irradiation studies carried out at BNL[8] with Amberlite IRN-77 in both H^+ and Na^+ forms, but only qualitative observations could be made. More recently, BNL[5] has attempted to determine in a quantitative manner the change in extent of agglomeration with absorbed dose. The use of a penetrometer to detect differences in depth of penetration of resin samples receiving different doses did not prove satisfactory due to inability to control other factors which affected the packing. Attempts to correlate absorbed dose with the water pressures required to move the resins when being sluiced, likewise yielded no quantitative measure of the agglomeration effect. With the IRN-77 cation resin,[8] qualitative observations were as follows: At 10^8 rad, agglomeration was not appreciable. At 3×10^8 rad, a definite stickiness was observed for both the H^+ and the Na^+ forms, with the H^+ form more affected. At 10^9 rad, both forms had a "gummy" appearance. The effect should not be deleterious in itself, but is an indication that damage to the resin has occurred.

Other indications of this kind are the darkening of color with increasing dose and physical breaking of resin beads. The BNL work[8] reported the progressive darkening of the cation exchanger IRN-77, and McFarland[6] did likewise for HCR-S cation resin. Beads of the latter resin are described as slightly darkened at 10^8 rad, and dark black after doses in the range 1 to 5×10^9 rad. Beads of the anion resin SBR-OH were observed to turn slighly brown, but apparently not very dark even at 10^9 rad. Beads of both the anion and the cation resin broke apart physically, as seen by microscopic observation.[6] The extent of breakage increased with increasing dose, and was more serious for the anion than the cation resin. Fracturing was observed with the anion beads at 10^8 rad, but not until 3×10^8 rad for the cation resin.

3.2.4.3 Effect on pH and Container Corrosion

Irradiation of a cation exchange resin in the H^+ form will cause loss of SO_3H^- functional groups and formation of SO_2 gas. As long as water is present, H_2SO_3 will form and dissociate to give hydrogen, bisulfite and sulfite ions according to the relations governing the respective ionization constants. In an oxidizing medium, a certain amount of sulfuric acid should also be formed. In any case, the water present will become more acidic as irradiation proceeds. If the resin is in a salt form rather than the H^+ form, acid will still be produced on irradiation, but the cation will act like a buffer in that some of it will be replaced by H^+ from solution so that the solution will be less acidic than one associated with an irradiated resin in the H^+ form.

These effects have been observed in experimental measurements made at BNL[8] and by Pillay at Penn State.[8] Results of irradiation at BNL of the nuclear grade cation exchange resin IRN-77 in the H^+ and Na^+ forms are presented in Table 3.4.

Table 3.4

pH of Water in Contact With
Irradiated Cation Exchange Resin IRN-77

Dose (rad)	pH	
	H^+ Form	Na^+ Form
0	3.5	6.8
10^7	2.5	4.7
1.25×10^7	2.5	
3×10^7	2.0	3.6
4.6×10^7	2.0	
10^8	1.5	2.9
3×10^8	1.0	2.1
10^9	0.6	1.3

Very high acidities were observed for the H^+ form, and only a little lower for the Na^+ form, at doses $>10^8$ rad. The amount of acid produced by the H^+ form was essentially linear with dose.

Results for anion resins irradiated by Pillay(7) and McFarland(6) show much smaller changes, although neither of them used the OH^- form of the resin. Pillay irradiated IRA-400 in the Cl^- form to 4.4×10^8 rad, and found it had the same pH (4.6) at the end of the irradiation as at the beginning. McFarland with Dow SBR-OH resin in the borate form observed a decrease of about 1 pH unit at around pH 8, on going from 10^8 rad to 10^9 rad.

Because of possible undesirable results from corrosion damage to metal containers of highly loaded resins, a considerable effort has been put into corrosion studies at BNL,(5,8) by Pillay(7) at Penn State, and by Marek and Rinker(6) at Georgia Institute of Technology. The general method used was to irradiate resins to various doses in contact with metal specimens, particularly mild and stainless steels, then examine the specimens, determine weight loss, etc. All three groups worked with cation resins; Pillay and Marek and Rinker also used anion resins. With the cation resins, appreciable corrosion of stainless steels was observed only at doses much $>10^8$ rad, whereas mild steel was affected more severely. The effects parallelled the pH changes observed on irradiation of the resin, as would be expected. Pillay found a certain amount of corrosion of mild steel with his anion resin, presumably at least partly because it was in the Cl^- form with a pH of 4.6. Marek and Rinker found no sign of corrosion on any stainless steel samples, even at the highest dose of 10^9 rad, again presumably partly because of pH, but this time a pH > 7.

3.2.5 Nuclear Grade Resins

Most of the work reviewed by Gangwer et al.[6] was done with ordinary commercially available resins, whereas the BNL work and McFarland's study described in the last section (3.2.4) used nuclear grade resins. The latter were used because of the wish to keep experimental conditions as close to plant conditions as possible, and nuclear grade resins appear to be used by most, if not all, nuclear power plants (see Section 2.1.1). The question then arises as to how comparable are experimental results obtained with these different grades of resins.

We have been unable to find published information on comparison of radiation effects in nuclear and non-nuclear grades of the same resin. However, a good indication of how comparable they will be should be gained by considering the known differences between them. These differences, which are not large in any case, are found mainly in the levels of impurities and the uniformity of bead size. Nuclear grade resins are typically limited to 0.5% fines smaller than 50 mesh and a similarly low percentage greater than 16 mesh. After the polymerization step in the manufacturing process, the material is treated to remove water-soluble organic compounds to a very low level. Inorganic impurities are kept low by special treatment during the manufacturing process. For Amberlite resins, these levels are: <200 ppm iron, <100 ppm copper, and <100 ppm heavy metals.

These particular specifications are aimed at achieving good operating parameters (e.g., low pressure drop) and maintaining the quality of the water in the liquid streams which have to be processed at nuclear plants. The most important consideration is to minimize corrosion of the reactor and turbine systems and radiation resistance would only be a consideration at all if, for some reason, it turned out to be poor for nuclear grade materials. Apparently, it has been assumed that there should be little difference between nuclear and non-nuclear grades of the same resin, and there seems to be no evidence to doubt that assumption.

Nuclear grade resins under irradiation would perhaps give slightly smaller amounts of gas and other decomposition products due to their smaller contents of soluble organic impurities. However, the organic impurity contents are quite low for both grades, so the effect could not be large. In any case, small (and therefore soluble) organic molecules are produced in both grades of resin by the action of radiation. As regards the inorganic impurities, in cation resins the salt forms with iron and copper as the bound cations have been empirically found to be much more stable to radiation than the H^+ form.[4] The reason for this was not given. In any event, the nuclear grade with its lower iron and copper impurities would probably be slightly less stable. However, again the difference would be very slight because the impurity levels are low in both grades. On balance, then, it appears that there should be very little difference between nuclear and non-nuclear grades of the same resin in terms of their resistance to radiation.

3.3 Choice of a Maximum Loading Limit

3.3.1 Basic Considerations

NRC has taken the position (in a draft BTP of October 30, 1981) that resins for disposal should not undergo degradation. Extensive degradation of organic ion exchange media would most certainly complicate the effective management of these wastes, as described above. However, we consider it fair to permit a small amount of degradation or damage if such damage will pose no problem to the burial site or the general environment.

Any dose of radiation will cause some damage to an ion exchange resin. Very small amounts of damage will be undetectable and obviously of no significance from a regulatory point of view. What is required is identification of a degree of damage which has a high probability of causing small but significant deterioration in performance and/or significant risk to the public health and safety. Radiation damage is always measured in terms of the dose received by the material damaged. Thus, in this section, the question is considered of what dose resins should be permitted to receive in order not to exceed the degree of damage described above. The question of whether to couch the Branch Technical Position in terms of a curie loading (Ci/ft^3 of resin) or a dose delivered to the resin is discussed in a later section (3.4).

We have taken the position (Section 3.2.5) that there is little difference in radiation stability between nuclear and non-nuclear grades of the same resin type. Accordingly, information from the literature on non-nuclear grade resins will be used along with more recent results of work on nuclear grade resins in order to arrive at a position on a maximum allowable dose to power plant resins for disposal (which will probably be only nuclear grade).

Another point which must be kept in mind is that the resins of most concern, i.e., those used for reactor coolant cleanup and which thus accumulate the highest RN loadings, are most likely to be mixed cation and anion, in the H^+ and OH^- forms. They may be mixed by the user, or purchased as mixed bed resin. The usual mixture at the plants responding to our questionnaire was a 1:1 ratio and this will be taken as the standard. The only types of resins considered are the strong base anion and strong acid cation polymerization types.

A final requirement for use of information from the literature is that this information pertain to resins with a similar water content to those disposed of by power plants. The latter are referred to as dewatered, and should have almost no free water in the container when shipped. A corresponding condition for resins used in the experimental work reviewed by Gangwer et al.[4] is termed "moist" or "swollen." Data obtained with dry resins and resins immersed in water cannot be considered acceptable for present purposes.

3.3.2 Minimum Deleterious Changes in Properties

We feel that it may be reasonable to permit a small degree of damage to resins for disposal; for example, the amount that would be caused by the dose

at which damage could just be positively identified. However, at that dose some other property might be degraded to a greater extent, perhaps too great to be considered permissible. Thus, the dose limit may have to be determined by the effect on the property most sensitive to radiation damage. Whether or not this will be necessary will depend on whether the deterioration in that property is capable of causing significant increased risk to the public, including operators at disposal sites. In this section the different properties affected by radiation are considered in the above context.

3.3.2.1 Gas Generation

The differences between McFarland's gas yields and those found in the earlier literature have been discussed (Section 3.2.4.1). The G-values calculated from McFarland's data at 7.9×10^8 rad were 0.09 and 0.69 for cation and anion exchanger, respectively. These values are 0.04 and 0.28 when calculated at 10^8 rad, which are somewhat lower than previous literature values. Since McFarland irradiated his resins in a dewatered state, using a dewatering procedure similar to that used in power plants, and since his experimental gas handling and measuring equipment was expressly designed for these experiments, we feel that his results must be given great weight. However, the conservative position requires consideration of the data indicating the greatest damage if the experimental work cannot be faulted. Thus, we have averaged the results from McFarland's experiments with the higher values of Mohorcic[16] on cation resin and Kazanjian[15] on anion resin, as given in Section 3.2.3, for purposes of calculating gas formation from representative power plant resin at two representative doses. McFarland's experimental data as given in Table 3.3 and the plots of Figure 3.2 are used in this calculation.

At a dose of 10^8 rad, McFarland's total gas production for a 1:1 cation: anion mixture amounts to 1.6×10^{-5} moles/g of resin, while at a dose of 2×10^8 rad it is 5.4×10^{-5} moles/g of resin. This does not include any correction for difference between H^+ and Na^+ form, since Mohorcic did not observe an appreciable difference between the H^+ and Li^+ forms of Dowex 50W for moist resin. Nor is it possible to apply a correction to the results for anion exchanger since there is no information on irradiation of borate form versus OH^- form. Kazanjian's result of 8.9×10^{-6} moles/g for Dowex-1, calculated from G-values given in Table 3.2, must be adjusted upward to account for the difference between moist and dry resin, and to convert from H_2 production to total gas production. For the latter, a factor of 1.8 is used, based on McFarland's results, and for the former, a factor of 2 is judged reasonable based on comparison with McFarland's results for moist anion resin. Mohorcic's result (Figure 3.1) has to be adjusted only for the difference between total gas and H_2 production, which is taken as a factor of 2.4 on the basis of McFarland's results. Kazanjian's result for anion resin is 3.6×10^{-5} moles/g of resin, and Mohorcic's value for cation resin is 2.2×10^{-5} moles/g of resin. Thus, for a 1:1 cation: anion mixture, the value taken is 2.9×10^{-5} moles/g. The average of this result with that of McFarland is 2.2×10^{-5} moles/g of resin at 10^8 rad. For 2×10^8 rad, the value for Mohorcic and Kazanjian is assumed to double to 5.8×10^{-5} moles/g of resin on the basis of the linear increase of yield with dose

obtained by Mohorcic. The average of this value with McFarland's then becomes 5.6×10^{-5} moles/g of resin for a dose of 2×10^{8} rad.

Assuming a sealed container, filled with dewatered resin to 90% of capacity, a resin density of 1 g/mL and a void volume of 50% in the resin, approximately 1.05 atmosphere of excess pressure would build up at 2×10^{8} rad based on the above average total gas yield value. In other words, the volume of additional gas produced would be somewhat greater than the void volume of the filled container. At 10^{8} rad the excess pressure build up would be 0.43 atmosphere. Although 1.05 atmosphere excess pressure would probably not damage the container, whether steel liner or high integrity polyethylene, the lower value would be preferable from the point of view of conservatism. Also, since gas pressure from bacterial degradation may build up in a sealed container, as reported by the Duane Arnold power plant (Section 2.1.5), it is particularly important not to generate appreciable amounts of gas by any other means. This leads to the choice of 10^{8} rad as the dose to be permitted for maximum loading based on gas generation.

3.3.2.2 Changes in Appearance

As pointed out in Section 3.2.4.2, the changes in appearance (swelling, darkening, agglomeration, and fracturing of resin beads) do not in themselves constitute effects which will produce a dangerous situation, but indicate that the resins have been altered, and have therefore undergone radiation damage. Resin shrinkage, occurring at doses much greater than 10^{8} rad, could release water and therefore should be avoided. Other changes in appearance become noticeable in the vicinity of 10^{8} rad. Fracturing of anion resin beads was observed by McFarland at 10^{8} rad but cation resin beads did not show the effect until 3×10^{8} rad. Since our reference resin is a 1:1 cation/anion mixture (Section 3.3.1), the lower dose must be chosen. It can be concluded then that the radiation damage indicated by these appearance changes is small, but definite at 10^{8} rad. Therefore 10^{8} rad is the maximum permissible dose indicated by these effects.

3.3.2.3 Exchange Capacity

This property is relatively sensitive to radiation. For resins irradiated to 10^{8} rad under the moisture conditions of dewatered spent resin, loss of exchange capacity is reported (Section 3.2.2) as 5-10% for cation exchangers and double that amount for anion exchangers. Quantitative data are not available for relevant resins at doses much below this, but it seems clear that detectable loss of exchange capacity would occur at half the dose, or 5×10^{7} rad, and probably even lower. However, loss of exchange capacity in itself does not consistitute a hazard to personnel or cause damage to containers. The resin damage it represents is removal of the functional groups, a process which does not damage the structural framework of the resin. Thus, it is felt that it is not appropriate to use it as the basis for establishing a limiting radiation dose.

3.3.2.4 pH and Corrosion

pH changes in cation resins in both the H^+ and salt forms are observed at quite low doses - 10^7 rad or less (Table 3.4). They are linked closely to loss from the resin of SO_3H^- functional groups. Anion resins in the salt form on the other hand, have shown only small changes at doses up to 10^9 rad. It would be expected that anion resins in the OH^- form would show greater changes, with an increase in basicity, due to generation of NH_3 and amines. We have found no information on irradiation of mixed cation and anion resins, but changes in pH of the mixtures could be much less than those seen for cation resins alone, due to takeup of SO_3H^- by the anion resin and neutralization of acid by released OH^-.

It appears that predicting damage to mixed resins by pH change on irradiation would be very uncertain, in that very large doses might show little or no pH change. This in itself is enough to negate its usefulness as a basis for setting a dose limit for resins. In addition, even if prediction were reasonably certain, the type of damage measured by pH change is largely loss of functional groups, and not damage to the structural framework of the resin. Finally, current experience at nuclear power plants indicates a wide range of pH values (2-9) is to be expected in undamaged spent resin. Thus, as with exchange capacity, we feel that pH change would not be appropriate as a basis for establishing a maximum permissible dose. These foregoing reasons also apply to corrosion effects, since corrosion is directly linked to pH.

3.3.3 Dose Rate Effects

The greatest difference between the experiments from which our radiation damage information has been obtained and the actual long term irradiation of spent power plant resins lies in the different dose rates in the two types of situations. A typical dose rate in the experimental work is 10^6 rad/h; it can vary from somewhat lower to 10^7 rad/h or higher. The spent resin, on the other hand, is subjected to a dose rate several orders of magnitude less. For example, resin loaded with Co-60 (5.3 yr half-life) in an amount which would deliver a total dose of 10^8 rad would be irradiated at considerably less than 10^3 rad/h. For the same total dose and Cs-137 as the source, because of the latter's 30 year half-life, the dose rate would be little more than 100 rad/h.

The differences in radiation effects which might result from such large differences in dose rate are not known. Recent work at Sandia[20] has shown that radiation at low dose rates (10^3-10^4 rad/h) can cause much more damage to polyethylene and polyvinyl chloride than the same total dose at the dose rates normally used in accelerated tests (10^6-10^7 rad/h). The mechanism proposed[20] involves peroxide formation and subsequent thermal decomposition, and it is not clear that such a mechanism would be operative with the largely aromatic structures of ion exchange resins. However in light of this work on aliphatic polymers, there remains the caveat that an increase in damage to ion exchange resins might result from the low dose rates encountered in practice. There is evidence (e.g., Swyler et al.[8]) that dose

rate variations of a factor of 10 have no effect on pH changes due to irradiation. However, the dose rates used are so much higher than those occurring with spent resins that no extrapolation can be made to the latter.

Intuitively, one could argue that in the spent resin situation, damage is done so slowly that some of it will have an opportunity to be repaired (e.g., long-lived radicals may be able to recombine). This would obviously not apply to processes like H_2 formation, where the intermediate H radical (H atom) is very reactive and short-lived. Gas buildup in a container, hydrogen buildup particularly, could however, be influenced by the long time frame dictated by a low dose rate. Over a period of many years, as gas pressure built up, unless the metal container were perfectly sealed, or if the container were plastic, the gas would gradually escape through small leaks or by diffusion through the plastic. This would constitute a mitigating effect in the case of a limiting dose to be set on the basis of gas generation. There is some mitigating influence in other damage effects as well (such as that causing container corrosion), because of the long time over which the radiation damage products which cause or enhance the effect are formed.

3.3.4 Selection of the Maximum Dose

A rationale for the choice of a maximum allowable dose to spent resins from nuclear power plants has been given in Section 3.3.1. In Section 3.3.2 the doses associated with the different effects caused by radiation were examined, and the doses identified at which small but detectable degrees of damage resulted. On the basis of all the information reviewed regarding the experimentally observed effects of radiation on resins of the same types as those used at nuclear power plants, in view of possible mitigating factors such as the very low dose rates involved in long-term irradiation, and taking into account the uncertainties involved in determining actual resin loadings at the plants, a maximum dose which power plant resins should receive is considered to be 10^8 rad.

It may be felt that a maximum permissible dose of 10^8 rad is too conservative, and indeed, because of uncertainties in measuring some of the radiation effects, a reasonable case can probably be made for setting the limit at some higher value, such as 2×10^8 rad. However, it should be noted that, in practice, determination of the RN loadings of large batches of used power plant resins is rather imprecise due to problems in obtaining representative samples. Thus, dose calculations based on such samples will also be imprecise. It is not inconceivable that a calculated dose might be a factor of 2 greater or less than that which the resin would actually receive, so that in an extreme case a batch might receive 2×10^8 rad when inaccurate analysis indicated it should receive only the limiting dose of 10^8 rad. It is considered that an adequate margin of safety would exist for resins inadvertently loaded so as to receive a total dose of 2×10^8 rad. It seems clear, however, that a 10^8 rad regulatory limit is not too conservative in view of the inherent imprecision in determining resin loadings encountered in actual plant experience.

3.4 Regulatory Use of Delivered Dose

3.4.1 Technical Basis

We feel that NRC's branch technical position (BTP) should be stated in terms of a delivered dose rather than a curie loading. It is technically imprecise to relate extent of damage to curie loading, since, for a given curie loading, different RNs (or different mixtures of RNs) can cause widely different degrees of damage. Our power plant survey showed that the RNs on resins for disposal vary from largely activation products to largely fission products. Co-60 is the controlling RN for activation products, and Cs-137 for fission products. For a uniform loading of 10 Ci/ft^3, the total dose delivered by Co-60 to a typical 100 ft^3 batch of resin would be approximately 6.4×10^7 rad depending on the exact shape of the resin container. For the same uniform loading of Cs-137 on a bed of the same geometry, the dose delivered would be 1.3×10^8 rad. The Sr,Y-90 couple does not appear to contribute much to the total activity in power plant wastes, but it should be noted that, at a loading of 10 Ci/ft^3, a total dose of 2.6×10^8 rad would be expected.

3.4.2 Calculation of Total Dose

Use of a curie level to assign a value for maximum loading might be permissible if calculation of total dose from a given loading of RNs was extremely difficult or subject to large error. However, neither is the case. Swyler, Barletta, and Davis[8] describe a method of calculating the dose, either at any time after loading or after total decay, from β,γ-emitting RNs. A full development of the equations is given in the appendix to their report.[8] For deposition of β-energy, the calculation can be made essentially exact, since the β-particle range is so short that >99% of the total β-energy is absorbed in the resin bed and the average β-energies are accurately known for the isotopes involved. For γ-energy deposition, estimation of the absorption of the γ-rays in the resin-water system is required. Values of the gamma ray constant, Γ, are given for a number of common RNs associated with the nuclear fuel cycle in the "Radiological Health Handbook."[21] Assuming tissue equivalency for the resin-water system, Swyler et al.[8] calculated the geometry factor, $\bar{g}$, given by Hine and Brownell[22] as a function of bed geometry. Estimation of $\bar{g}$ involves the greatest uncertainty in the γ-dose calculation, but it is well within the limits that would be required for normal reporting of loadings to burial sites.

The calculations are not difficult and could be done routinely by the generator. Indications from the survey are that some generators use calculations based on radiation field (similar to dose rate) to determine their resin loadings. Such calculations have similar requirements and assumptions to those involved in the calculation of Swyler et al. A computer program based on the latter has been written and is given in Appendix E.

3.4.3 Sr-90 Reporting Requirements

The results of our power plant survey indicated that rather small amounts of Sr-90 are present in their resin wastes. Only 3 plants reported it in their list of principal RNs on dewatered resin and only two gave a value for its level on spent resin. One of the latter plants routinely used a value of 1% of the Cs-137 level, since periodic analyses had never given a higher result. The other plant sent a recent analysis showing the Sr-90 content of the resin as 2.5% that of Cs-137. Based on these results and on phone calls to several other plants, it appears that Sr-90 levels in the liquid streams and on the spent resins are in general much lower than Cs-137 levels. In such cases, it would therefore not need to be considered for calculating long-term dose to the resins, particularly if activation products constitute the bulk of the activity disposed of. However, due to the high dose from the Sr,Y-90 couple compared with that from the other major contributors, (twice that of Cs-137 and four times that of Co-60 for large volumes of resin--see Section 3.4.1) we feel it is prudent to require that generators report Sr-90 levels on their coolant cleanup resins to disposal sites. Further, they should explicitly include Sr,Y-90 contribution in their total dose calculation.

3.5 Recommendations for Additional Research*

It has been pointed out[4] that there is a certain lack of agreement, as well as a lack of quantitative data, in the published literature on radiation effects in organic ion exchange resins. This is particularly true for anion resins. In attempting to arrive at a quantitative value for maximum permissible dose to resins being disposed of by nuclear power plants, we had to make several extrapolations from one form of resin to another (e.g., Li^+ form to H^+ form), and from dry to moist resin, because the required data were not available. In order to avoid uncertainties of this kind, a satisfactory data base should be developed, particularly for resins of the types used in power plants. The following recommendations for additional research, if implemented, would provide much of the information needed. It should be noted that moisture content is critical to the results obtained. Thus, in all the experimental work suggested below, the water content of the resin samples must be accurately known.

- To permit use of information from earlier literature obtained with non-nuclear grade resins, a check should be made comparing samples of nuclear grade and non-nuclear grade of the same resin, both anion and cation. Properties to be used for comparison are gas generation (preferably individual gases), changes of pH, and loss of exchange

*Since this report was published in draft form (BNL-NUREG-30668, January 1982) experimental work has been performed at BNL in most of the areas suggested. Results are given in the quarterly progress reports, "Characterization of TMI-Type Wastes and Solid Products," BNL-NUREG-51499, April 1982, BNL-NUREG-31413, June 1982, and BNL-NUREG-31568, July 1982.

capacity. Dewatered resins should be used, at least in the H^+ and OH^- forms and absorbed doses should be adequately distributed over the range 10^7 rad to at least 2 to 3 x 10^8 rad.

- Continuing from this point with nuclear grade resins only, the same properties should be determined for salt forms, such as lithiated and borated, which are used in power plants. Both dry resins and dewatered resins should be tested, in both the salt forms and the H^+ and OH^- forms.

- There is almost no information on irradiation of mixed anion and cation resins. This situation should be remedied in order to provide data relating to the conditions in actual plant use. Irradiations should be carried out, over the same dose range as before, on samples of 1:1 anion: cation resin mixtures in the H^+/OH^- form and in both the dry and dewatered state. The dewatered samples should be tested for pH change in particular. It is thought that gas generation from mixed resins should be the sum of the amounts generated from the cation and anion resin irradiated separately, and this point should be checked.

3.6 Dose From Short-Lived RNs During Plant Operation

3.6.1 Potential for Damage

The possibility that decay of the short-lived RNs on resins in demineralizer systems during reactor operation could deliver a significant dose to the resins was pointed out in Section 2.2. In that section, analytical results provided by several power plants for their reactor coolant were given (Tables 2.4-2.8) along with data on gross β, γ-activity in the feed to the coolant cleanup demineralizers (Table 2.2).

The analyses indicate that reactor coolant routinely contains relatively high activity levels of short-lived RNs (several days or less) and low levels of the longer-lived isotopes (such as Co-60 and Cs-137, with half-lives, respectively of 5.3 and 30 years) which make up the bulk of the activity on spent resin for disposal. The high overall DFs reported for gross β,γ-activity across the beds [which agree with those given by Lin[1]] show that while the beds are in service, they are continually receiving an appreciable radiation dose from the short-lived nuclides, since the activity loadings are of the order of 1 mCi/mL of resin (28 Ci/ft^3).

3.6.2 Dose Determinations and Relevance to a Maximum Loading Rule

In order to determine whether such a dose would contribute importantly to the overall dose eventually received by the resin, calculation of its magnitude at Dresden Unit 2 was carried out, based on the extensive analytical data and information on plant operation supplied in their response to the questionnaire and by subsequent telephone contact. Of the RNs they monitor

(see Table 2.4), those that deliver the bulk of the short-term dose are I-131,132,133,134,135, Ba,La-140, Cs-138, Na-24, Fe-59, Zr-95, Ba-139, Tc-104, and Ce-141. Their mean lives range from less than an hour to 3 months.

Since in 5 mean lives an isotope will build up to within 1% of its saturation level on a fresh bed, those isotopes with mean lives of 20 days or less will reach their saturation levels in 4 months or less. From the time they reach saturation, their rates of decay are equal to their rates of build-up on the resin, assuming constant coolant flow rate through the bed and constant reactor power. Thus, they deliver a continuous dose to the resin all the time the bed is in service. After the bed is removed from service and the RNs decay, the dose from their decay will exactly compensate for the deficit in dose during buildup, before saturation is reached. Hence, the total dose delivered by the short-lived isotopes is simply the dose rate at saturation multiplied by the time the bed is in service.

For isotopes of slightly longer mean life (τ) which do not quite reach saturation in 6 months, the dose delivered by decay after removal of the bed from service will not be quite enough to compensate for the difference between actual and saturation levels during buildup. However, for an isotope of mean life 90 days (6 months $\simeq 2\tau$) the error involved in assuming complete compensation is only about 1%. Thus, Zr-95 (τ = 94 days) and Co-58 (τ = 102 days) can be included in the calculations with introduction of an insignificant error in the total dose from all short-lived isotopes.

In Appendix F, calculations are given for the β-dose from Ba-140 using the above treatment and a treatment based on the method of Swyler et al.[8] described in section 3.4.2, which gives the same result. This second treatment is used to determine the γ-dose from Ba-140. The sum of the β- and γ-doses from Ba-140 is calculated as 1.3×10^6 rad. Applying the same methods to the other short-lived isotopes listed above leads to a total delivered dose of 6.4×10^6 rad in 6 months of operation (plus subsequent decay time).

This calculated dose is an underestimate for several reasons. There are a number of fission product isotopes of high fission yield (e.g., Rb-88,89, Y-92,93,94, Ba-142, Te-133,134) which will probably also be present but which are not measured by the analytical methods employed by this particular plant. Also, in the calculation dose is assumed to be equally distributed between water and resin in the bed. This is a good approximation for γ-dose, but β-dose will be greater to the resin. A realistic estimate of the total dose delivered at Dresden by all short-lived isotopes is most likely $\lesssim 10^7$ rad. The Dresden situation is probably average, since the fission product activity in the coolant comes from uranium "plated out" on core surfaces rather than from fuel element leaks. Some plants have lower activities in the coolant and/or change beds* more frequently so that their resins receive a lower dose. Other plants operate from time to time with slight fuel leaks, so that

*"Bed" as used here includes powdered demineralizers.

fission product concentrations in the coolant could be higher, but their coolant cleanup beds* may have much shorter service times than Dresden's, and thus radiation dose to the resin during bed operation probably will be comparable to Dresden's or $\leq 10^7$ rad.

We conclude that it is unlikely for any power plants to produce waste resins receiving a dose from short-lived isotopes much $>10^7$ rad, or 10% of the 10^8 rad suggested as the maximum permissible limit. Dose from this source, therefore, would normally contribute an amount which would be within the limits of error for estimating a dose of the order of 10^8 rad. The significance of this "extra" dose, however, is that it supports a conservative position for setting a maximum permissible dose from long-lived RNs, since an appreciable uncertain contribution to total dose will always occur from short-lived RNs, and perhaps from other sources of radiation we have not considered.

3.7 Solidified Resins

It is beyond the scope of this report to consider dose limits for solidified resins. However, it is appropriate to draw attention to recent studies of radiation damage to cement-resin composites which indicate a high degree of stability for certain solidified resin forms.(5) In view of such results, a dose limit of the order of 10^9 rad to the resin might be considered for suitable solidified forms which could lead to significant waste volume reduction of benefit to both the generator and the disposal site. The results of these experiments and recommendations for additional research in this area are discussed below.

3.7.1 Experimental Results of Radiation Damage Studies

The manner in which p operties of EPICOR-II type ion exchange media changed with radiation exposure has recently been investigated.(5) The material studied was a proprietary mixture called D-mix, consisting mainly of organic resins and claimed by the vendor to be representative of the Epicor-II first-stage liner material. Irradiations of D-mix alone were carried out to determine its effect on gas generation, agglomeration, pH, and corrosion of potential container materials. A number of irradiations of D-mix solidified with cement were also carried out, and the results of the testing done on these composites as they relate to the matter of a maximum permissible loading, are summarized below.

- Leachability of solidified D-mix/cement composites was not increased by irradiation to a total dose of 10^9 rad.

- No deleterious effects to the mechanical integrity of the composites were observed, as measured by tensile splitting strength.

*"Bed" as used here includes powdered demineralizers.

- There was no measurable corrosion of stainless steel either with D-mix alone or with composites at any dose.

- No unambiguous evidence of radiation enhancement of mild steel corrosion was found for samples in contact with D-mix/cement composites at a total dose of 10^9 rad. This is in contrast to the enhanced corrosion observed with D-mix alone, which was noticeable at 10^8 rad.

- The major gas produced as a result of irradiation of the composites was hydrogen. Very little CO_2 or methane were formed in the composites, whereas appreciable quantities were generated in D-mix alone.

- As described in Section 3.2.4.1, with both D-mix alone and D-mix/cement composites, a marked decrease in oxygen content of the atmosphere in sealed containers upon irradiation was observed (nearly complete depletion in the case of the composites).

3.7.2 Conclusions Regarding Maximum Loading

From the results given above, it can be concluded that mechanical integrity and leachability of the D-mix/cement composites are not affected by irradiation to a total dose of 10^9 rad. There may be a slight enhancement of corrosion of mild steel at the higher doses, but this is not definite. Thus, from the point of view of these properties, the composite waste form could be used for incorporation of D-mix which would deliver such doses. Gases such as CO_2 and light hydrocarbons are formed in only small amounts, suggesting that degradation of the organic resins present in D-mix may be less extensive upon solidification with cement. However, gas generation (mostly hydrogen) is still appreciable from the composites. Based on all these results with the single material, D-mix, it appears that solidifying resins with cement would permit a loading which would give a dose to the resin, if not solidified, of considerably higher than 10^8 rad, and perhaps as high as 10^9 rad. A similar conclusion is reached for resins solidified in Dow Chemical Company's vinyl ester-styrene polymer according to a study by Dow using both BWR and PWR resin wastes.[23] However, insufficient data are presently available on which to base a recommendation to permit higher doses such as 10^9 rad to composites, and in the next section suggestions are given for experiments which could generate the required data.

3.7.3 Recommendations for Additional Research*

The principal requirements to be met by additional research are to generate results with material that is not proprietary, and to broaden the information base with respect to both resins and solidification agents. Some specific suggestions are:

- Cement composites should be prepared and tested using commercially available, non-proprietary, anion and cation exchangers, individually and in a 1:1 mixture. Nuclear grade resins should be used.

- The unsolidified resins should be tested at least for gas generation, to provide a reference point for comparison with the composites.

- A range of solidification agents consistent with those currently in use at nuclear power plants should be tested. An organic material such as Dow vinyl ester-styrene polymer should be compared with inorganic solidification agents such as cement and cement-silicate mixtures.

- The resin/waste composites should be tested at least for gas generation, changes in leachability, and mechanical stability, to total doses of 10^9 rad. Leachability and mechanical stability tests used should be those described in the draft Branch Technical Position.

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*Since this report was published in draft form (BNL-NUREG-30668, January 1982), experimental work has been initiated at BNL in the first area recommended, and work in additional areas is planned. Preliminary results are given in the quarterly progress report, "Characterization of TMI-Type Wastes and Solid Products," BNL-NUREG-31568, July 1982.

4. TEST FOR PERMITTING EXCEPTIONS TO THE GENERIC RULE

There is a possibility that a generator may wish to use a resin, or resin mixture, which it claims can be loaded to a level higher than that allowed by the BTP, without resulting in degradation. Examples exist of resins which have considerably greater radiation stability than those presently in general use, particularly the anion resins with pyridine units in their structural framework. NRC's draft BTP requires that a generator wishing to load a resin to some higher level than permitted by the rule must demonstrate that the resin will not undergo degradation at that loading level.

In developing a suitable test to provide this demonstration, several conditions should be met:

1. The test method should adequately simulate the chemical and radiological conditions which would be encountered in actual practice.

2. The test should be reasonably simple and consistent with the types of operation and experimental equipment required to obtain accurate results. Simplicity, however, cannot be considered a limiting factor, since this would be a "one time only" test, and not a routine procedure.

4.1 Adequate Simulation of Conditions Expected in Practice

4.1.1 Use of Chemical Solutions

It has been argued that the effects of radiation on resins, either in water or containing water, can be produced by treating them with chemical solutions, particularly solutions of H_2O_2. It is undoubtedly fair to say that some of the effects of radiation will be produced by such solutions, since H_2O_2 is inevitably formed by radiolysis of water. However, it would be a gross oversimplification to contend that this or any other chemical can adequately simulate exposure to radiation at the doses required in a test.

In the first place, H_2O_2 cannot cause ionization. Aside from this obvious difference, in fact because of it, a large chemical discrepancy also exists. Ionizing radiation produces relatively high concentrations of very reactive radicals (e.g., $H^{\bullet}$, $HO^{\bullet}$, and $HO_2^{\bullet}$) as well as H_2O_2, by radiolysis of the water inside the resin beads. All these species are thus able to attack all parts of the resin structure throughout the bead. In the case of dewatered resin treated with H_2O_2 solution, on the other hand, the radicals will not be present at all and the H_2O_2 will be able to attack only the outer bead surface until diffusion into the interior can take place. Another important chemical species which is missing from the chemical H_2O_2 system is hydrogen gas which is produced from both resin and water by radiation.

A marked difference between the radiation and purely chemical systems is the manner of attack on the organic resin. None of the chemical bonds in the

resin polymer molecules are immune to radiation. Most of the radiation damage begins with direct scission of C-C and C-H bonds, and the C-S and C-N bonds to the functional groups, to form free radicals of all sizes, from H atoms to radicals of "molecular" weights of many thousands. These radicals are then available for reactions with each other and with all the mobile species (H_2O_2 and radicals) formed from water radiolysis. In the H_2O_2 chemical system, there is apparently a certain amount of C-C bond attack, particularly when iron and/or copper ions are present to act as catalysts, but this attack is slow, with relatively low H_2O_2 concentrations. H_2O_2 attacks C-H bonds in oxidation reactions, whereas in radiation systems abstraction of H atoms by H·radicals to form H_2 is a major contributor to the overall reaction.

It must therefore be concluded that use of chemical solutions cannot adequately simulate the chemical conditions, let alone the radiological conditions, specified in the draft BTP.

4.1.2 Use of Radiation

From the foregoing discussion it becomes obvious that the only way to simulate radiological conditions, which will in the process simulate chemical conditions, is to irradiate the test sample with radiation of low LET similar to the β,γ-irradiation given by radionuclides to spent resins in practice. This can still only simulate actual radiation conditions, not reproduce them. There is no way to reproduce the low dose rates to resins at burial sites without carrying out experimental irradiations for many years. This matter of dose rate differences has been discussed at length in Section 3.3.3.

4.2 Choice of Irradiation Method

Irradiation can be carried out either internally, by adding β,γ-radioactivity to the resin sample, or externally, by providing the radiation dose from an external source such as an electron accelerator or an X-ray or γ-source. Nuclear reactor irradiation is not suitable because the radiation provided has a large component of high LET radiation, i.e., energetic protons produced by collision of fast neutrons with hydrogen atoms in water and resin. Determination of absorbed dose in reactor irradiations is also extremely difficult.

4.2.1 Internal Irradiation

Internal irradiation would most closely simulate spent resin conditions. It could be carried out by loading a resin sample with the appropriate amount of a relatively short half-life isotope to give the resin the required dose upon essentially complete decay. The half-life would have to be short so that the radioactivity on the resin sample could decay essentially completely (at least 10 half-lives) before the resin was tested, and so that the total time for the whole procedure was kept reasonable. A pure β-emitter would be preferable for this, since total dose could be determined quite accurately. Estimation of the γ-dose from a β,γ-emitter would be subject to appreciable error for a small sample. However, there is a major disadvantage to use of internal radiation, even

assuming an appropriate RN source could be found, and that is the complexity of working at least behind shielding, and perhaps in a hot cell, coupled with associated worker exposure.

4.2.2 External Irradiation

External irradiation has several advantages over internal. One is that dosimetry can be made quite accurate, and dose rates under various conditions are determined regularly at most electron accelerators, and γ-ray and X-ray irradiators. Another is that irradiations can be carried out with essentially no exposure to personnel. Still another is that there would be no radioactivity in the sample, whereas using an internal source, there might be residual radioactivity in the sample (either from the source isotope or an impurity) when the sample was being tested. From the technical point of view, external irradiation appears to be the preferred method. Also, it is felt that such an irradiation would be easier to monitor by a regulatory agency, and would likely be preferred by a power plant, since service irradiations can readily be arranged and are not expensive.

Regarding the choice between fast electron irradiation and X-ray or γ-irradiation, it is concluded that X-ray or γ-irradiation is preferable. For electron irradiation, the sample geometry would be crucial if anything like a uniform irradiation dose were to be achieved. The irradiation cell would also have to have a thin window to permit the electron beam to reach the sample. Neither of these constraints applies to X-ray and γ-irradiation. With both types a small sample will obtain a uniform radiation dose, and the cell wall thickness and cell construction are not critical.

4.3 Suitable Properties on Which to Base a Test

An essential criterion for choosing a property of the resin as a basis for a damage test is that the property must be one which would be possessed by any conceivable organic ion exchange resin that a generator might propose as a candidate. Since any resin proposed will be organic and will contain functional groups, it will generate gas due to absorption of radiation, and will lose functional groups, and thus exchange capacity. It may or may not possess the other properties which we have evaluated. The various properties are considered below in terms of the essential criterion and other bases for judging suitability.

4.3.1 Changes in Appearance

As pointed out in Sections 3.2.4.2, most changes in appearance, such as darkening of color, bead fracturing, and agglomeration, have been related to radiation dose in only a qualitative way, because of inability to make quantitative determinations of the effect. In the case of swelling, where quantitative determinations can be made, the degree of swelling is a function both of percent cross-linking and of the concentration of the functional groups, which are hydrophylic. Under irradiation, the cross-linking and exchange capacity vary in a complex way so that swelling may occur at low dose and the effect

reverse at higher doses. Thus, depending on the resin, for a given dose one might observe swelling, no change, or shrinkage.

The first three effects can be ruled out as the basis for a test because of the present inability to make quantitative determinations of their magnitude. Swelling would not necessarily meet the essential criterion, although it is difficult to envision an organic resin that would not exhibit some sort of swelling effect. In any case, it is judged not to be a good property on which to base a test because of the difficulty in knowing what level of resin damage goes along with what degree of volume change and therefore what the allowable level should be. With further work and a better understanding of the factors influencing this property, a suitable test could probably be developed. However, on the basis of the information in the literature, it is not presently acceptable.

4.3.2 pH Change and Enhancement of Corrosion

pH change cannot be considered a suitable effect on which to base a test for reasons similar to those applying to resin swelling, i.e., inability to know at what level to set the allowable limit. The usual practice in power plants is to use a mixed bed of cation and anion resins, generally 1:1 cation:anion, for coolant cleanup. Irradiation of such a mixture appears to give only small pH changes after quite large doses, although fairly large changes occur with strong acid cation exchangers alone. Thus considerable damage could be done to a resin mixture before a properly measurable change in pH occurred (giving the illusion of great stability) because the effect is not additive, i.e., the pH change of the mixture is not the sum of the changes for cation and anion exchangers separately.

Since enhancement of corrosion depends on pH, the conclusion must be that this effect cannot be the basis for a test as long as pH change is considered unsuitable. In any case, because of the nature of the corrosion process, enhancement of corrosion appears to be the least straightforward property to apply in a quantitative manner, aside entirely from any uncertainties regarding pH changes.

4.3.3 Exchange Capacity

It has already been pointed out that exchange capacity meets the essential criterion of being applicable to any potential resin candidate. It is a direct measure of the number of functional groups present, and loss of functional groups cause loss of exchange capacity. It was not deemed appropriate to base a maximum allowable dose on the behavior of this property, in that the loss of functional groups does not represent damage to the structural framework of the resin (Section 3.3.2.4). This, of course, has no bearing on its use in a standard test. Such a test should be completely suitable for this purpose in terms both of direct relationship between dose and measured effect, and of simplicity in the procedure. In this connection for example, only simple wet chemical methods would be required, and samples could probably be irradiated in air. We feel, however, that a test more closely related to degradation of resin structure would be more appropriate, if a suitable one is available.

4.3.4 Gas Generation

Gas generation also satisfies the essential criterion, in that any conceivable candidate resin will generate gas when irradiated. Hydrogen will always be generated, and methane also seems to be formed from both cation and anion resins, though in much smaller amounts. CO_2 and CO are also formed from both types of resin. There seems to be no purpose in basing a test on measurement of only one gas, since that would require analysis, an extra step with additional chance of introducing errors into the measurement. Thus, from the point of view of both simplicity and accuracy, total gas should be measured.

The formation of both total gas and the major constituents (H_2, CO_2, CO) has been found to be reasonalby linear with dose (Section 3.2.3 and Figure 3.1), although McFarland in recent work has reported evidence of a dose threshold in the formation of total gas, and a slight increase in the slope of the yield-dose curve with increasing dose (Section 3.2.4.1 and Figure 3.2). This does not affect the choice of gas generation for purposes of a test, since a limiting value for allowable gas production can still be set when the production rate departs slightly from linearity. Also there is no problem with use of a mixture of anion and cation resins, since the gas production of the mixture is the sum of the amounts produced from irradiation of the individual resins.

In terms of simplicity of measurement, gas generation should also be suitable. Measurement of the amount of gas produced, although requiring only standard high vacuum equipment, will demand considerable care in making the experimental observations. This is not judged to be a difficult requirement to meet, and from all points of view, gas generation is felt to be completely suitable as the basis for a standard test.

4.3.5 Final Choice of Measurment to be Used

From the foregoing discussion of suitability of the various properties affected by radiation, the choice of property on which to base a test narrows down to exchange capacity and gas generation. Since it is felt that exchange capacity should be the basis only if no other property is suitable, and gas generation is judged completely suitable, we recommend that measurement of gas generated be used in the standard test for permitting exceptions to the maximum loading rule.

4.4 Development of the Test

A number of factors must be taken into account in developing a test procedure of this nature. In the following section we discuss those we feel are either critical, or at least important.

4.4.1 Specific Requirements

1. An important requirement is that the resin be irradiated dry. This requirement has a two-fold purpose. First, it provides a known

standard form for the starting material without having to comply with the requirement of some arbitrary water content, and second, it avoids complications from formation of gas by radiolysis of water.

2. In connection with a standard form, a further requirement is that resins to be tested must be in the H^+/OH^- form rather than in salt form, since there can be considerable variation in gas yields between different forms.

3. In order to avoid possible interaction of the resin sample with metal in a metal irradiation cell, the sample must be contained in a glass or quartz holder inside the cell. Use of a Pyrex irradiation cell avoids the problem.

4. Sample size must be large enough to produce an accurately measurable amount of gas at the radiation dose the resin is designed to accept without degradation. Some representative figures are considerd in the next section (4.4.2) which deals with the allowable limit for gas production.

5. The radiation source must be either an X-ray or γ-source to provide sufficiently penetrating radiation. With a fast electron source, a thin window would be required to permit the electrons to reach the sample. This poses problems in cell construction and in carrying out the irradiation in such a way as to provide a reasonably uniform dose to the sample, especially since the sample must be reasonably large. The thin window, particularly if of large area, would be a potential problem in terms of rupture under vacuum or above atmospheric pressure. With X-ray or γ-irradiation, sample geometry and size, and cell construction are not problems.

4.4.2 Limit for Acceptable Amount of Gas Generation During Test

The maximum amount of gas which can be generated by a candidate resin being proposed for exception to the maximum loading rule, has to be based on the amount considered normal for a dose of 10^8 rad to the typical nuclear grade resins used by power plants. The reference resin is a 1:1 cation:anion mixture in the H^+/OH^- form as typically used for reactor coolant cleanup.

The only study on irradiation of cation resins which produced quantitative data on gas generation from dry resin in the H^+ form appears to be that of Mohorcic and Kramer.[16] Their value for H_2 production was 2.2 x 10^{-6} moles/g of resin at 10^8 rad, with CO and CO_2 contributing another 3.5 x 10^{-6} moles/g, and SO_2 7.1 x 10^{-6} moles/g, for a total of 1.3 x 10^{-5} moles/g. The SO_2 value seems high in relation to H_2 yield, since McFarland[6] reported only a very small amount of SO_2 from his dewatered or moist cation resin. However, in McFarland's experiments, SO_2 may have been retained in solution in the water in the resin matrix. Mohorcic irradiated his dry resin in an evacuated glass vial, the conditions to be used in our test, which should have allowed SO_2 to be liberated. He also analyzed the generated

gases by mass spectrometer, and it is difficult to see how he could confuse mass peaks from SO_2 with those of the other gases. For these reasons, we feel that his gas yields for that particular resin (Dowex 50W) must be accepted, and we therefore take the value of 1.3×10^{-5} moles/g of resin as the total gas yield for a reference cation resin in the H^+ form.

For the anion resin, the results of Kazanjian and Horrel[15] give a value of 9.3×10^{-6} moles of H_2/g of resin at 10^8 rad for dry Dowex-1 in the nitrate (NO_3^-) form. This compares with McFarland's values of 1.5×10^{-5} moles of H_2/g and 2.8×10^{-5} moles of total gas/g for dewatered or moist Dow SBR-OH in the borate form, and shows about the expected difference between moist and dry resin. Accordingly, Kazanjian's value is considered acceptable. There is so little quantitative information available on anion resins that it is not clear what to expect for the difference between dry OH^- and dry NO_3^- forms. The salt form is perhaps more stable, but in the absence of definite evidence to that effect, we feel the same value should be used for the dry OH^- form as for the dry NO_3^-. Then, using the ratio of H_2 to total gas of 0.54 obtained by McFarland leads to a value for total gas of 1.7×10^{-5} moles of gas/g of resin for a reference anion resin in the OH^- form.

Based on the above yield values chosen for the reference individual anion and cation resins, the total gas yield for the reference 1:1 anion: cation mixture is 1.5×10^{-5} moles of gas/g of resin at 10^8 rad. The figure is bound to be somewhat arbitrary, due to the uncertainties of several of the assumptions, but is certainly in the right range. In terms of volume at STP, this value would be 0.3 cc of gas/g of resin.

4.4.3 Test Procedure

1. Sample Irradiation Cells

The irradiation cells can be constructed from Pyrex tubing of appropriate diameter. They must be large enough to hold approximately 20 cm^3 samples of resins, but total void volume must be kept low so that the pressure of gas produced can be measured as accurately as possible. A side arm containing a break seal must be attached above the level to which the cell will be filled with sample. This will be used for connecting the cell to the gas measuring system after irradiation. The tubulation at the top of the cell should be of appropriate diameter for attaching to a vacuum system and for making a constriction for sealing off the evacuated cell. Pyrex irradiation cells of this type have recently been described.*

Metal irradiation cells can be used, but require special parts, such as all metal valves and fittings, since no organic gaskets, O-rings, valve packings, etc., can be placed in the irradiation field. Radiation damage to the

*K. J. Swyler and R. E. Barletta, "Irradiation of Zeolite Ion-Exchange Media," Draft Report, BNL-NUREG-30631, December 1981.

organic components could cause loss of vacuum by leaking at the doses to which the resin samples are to be irradiated. If metal cells are used, the resin samples should be placed in glass or quartz liners inside the cells to avoid interaction of resin with the metal. The steps of the procedures are written in terms of glass cells. If metal cells are used the steps will be essentially the same, except that metal valves will be used in place of glass break seals and seals at constrictions.

2. Sample Preparation

a. The resin must be in the H^+ form if cation exchanger, OH^- form if anion exchanger, or H^+/OH^- form if mixed cation/anion.

b. Thoroughly dry the resin by heating it at 110°C for 24 hours.

c. Weigh out, in a dry atmosphere, six samples of approximately 10 g each and place each sample in a separate Pyrex sample irradiation container.

d. Alternatively, in place of steps b and c, six samples of undried resin can be taken, of such a size that when dry, they will weigh approximately 10 g each. Place each in a separate, weighed Pyrex irradiation cell and dry as in step b. Weigh the cells again to determine the weights of dry resin.

e. Attach the Pyrex cell to a glass vacuum system capable of use for volume calibration (the gas measurement system would be appropriate), and constrict the connecting tubulation so that it can be sealed under vacuum.

f. Evacuate the cell and determine its void volume up to the constriction using dry N_2 or other inert gas.

g. Outgas the sample and seal off the cell, under vacuum, at the constriction.

3. Irradiation

Place the irradiation cell in a position in the X-ray or gamma-ray irradiation facility where the dose has been accurately determined, and leave it until the desired dose has been reached. Three samples are to be irradiated to 10^8 rad total dose, and three samples to the dose the generator wishes to use to qualify his resin for an exception. An empty sample cell for use as a blank is also to be irradiated to the higher dose. The purpose of irradiating samples to 10^8 rad is to provide comparison with other resins, and to provide a further measure of confidence in the results at the higher dose.

4. Measurement of Gas Produced

a. The gas measurement system must contain a calibrated volume, which includes the pressure measuring device, similar in magnitude to that

of the void volume of the samples. The pressure measuring device could be a mercury manometer with adjustable level on the gas measuring side to permit the volume to be kept constant. Other acceptable pressure measuring devices include a small volume metal vacuum guage or pressure transducer, reading in mm.

b. After irradiation, attach the side arm of the sample (or blank) irradiation cell to the gas measurement system with breaker in place.

c. Evacuate throughly and check for leaks. When the whole system is sound and evacuated, break the break seal and measure the pressure in mm in the whole system (irradiation cell plus gas measuring system).

d. Calculate the specific amount of gas generated during irradiation of the resin according to the formula

$$V_G = \frac{(V_m + V_c)}{m_R} \cdot \frac{P_s}{760} \cdot \frac{273}{T_s}$$

where V_G is the volume of gas produced at STP per g of dry resin.

V_M is the volume of the measuring system

V_c is the void volume of the irradiation cell

m_R is the weight of dry resin

P_s is the pressure in mm in the whole system

T_s is the temperature of the system in K.

e. In order to qualify the resin for use at the higher dose used in the test irradation, the value of V_G must not exceed 0.3 cc of gas at STP/g of dry resin.

5. SUMMARY AND CONCLUSIONS

5.1 Power Plant Survey

The resin usage questionnaire was returned by 23 power plants. The principal results of interest to this report are given below under several appropriate headings.

1. Resin Usage

   - No plants reported use of inorganic ion exchangers.
   - Strong acid cation resins and strong base anion resins of the polymerization type are the kinds in general use.
   - Nuclear grade resins were used by most of those plants responding.
   - The resins used for cleanup of reactor coolant by most plants were mixed anion and cation, usually in a 1:1 ratio, and in the H^+/OH^- form.
   - No evidence of radiation damage to the resins was reported.

2. Radioactivity in Liquid Streams

   - The highest radioactivity levels in liquid streams were reported in the reactor coolant and boron recycle. They were 10-100 times higher than those in liquid radwaste and in the streams from spent fuel pools.
   - Essentially all the activity in the reactor coolant is due to short-lived RNs. The longer lived fission and activation products, Cs-134, Cs-137 and Co-60, are present at very low concentrations.
   - There was considerable variation in the coolant RNs reported from different plants, however noble gas isotopes and I-132 through 135 almost always contribute the bulk of the activity.
   - Levels of activity in the coolant at several plants were high enough that a significant dose could be delivered to the coolant cleanup resin from steady-state concentrations of the short-lived RNs sorbed or trapped by the bed.

3. RN Loadings on Resins for Disposal

   - Spent resin from coolant cleanup generally had much higher RN loadings than resins from other demineralizer systems.

- There was a wide variation in the RN loadings on coolant cleanup resin reported by the different plants. Typical loadings ranged from 0.1 to 30 Ci/ft^3, while maximum loadings ranged from 0.3 to 60 Ci/ft^3 of isotopes with half-lives longer than 5 years. Both BWRs and PWRs showed this variation. The BWR range for maximum loading was 0.3 to 60 Ci/ft^3 and the PWR range was 1 to 40 Ci/ft^3.

- Five of the reporting plants have shipped resins loaded to >10 Ci/ft^3 and several others have produced spent resin loaded to >10 Ci/ft^3. Typical loadings were said to be less than maximum. Three of the plants reporting loadings >10 Ci/ft^3 on spent resin solidify before shipping, so the curie level of their shipped waste form would be less than that on the spent resin itself.

- The distribution of the relatively long-lived RNs varied from >90% activation products (Co-60, Mn-54, etc.) at some plants to >90% fission products (Cs-134, 137) at other plants.

- Sr-90 appears to be a minor constituent of the RNs on spent resins. It was mentioned by only three plants as being present. Only one plant gave an analytical result, and that was 2.5% of the Cs-137. Personnel contacted at other plants stated that their Sr-90 was low; at one plant it had never been as high as 1% of the Cs-137.

- Several plants reported TRU isotopes on spent resins, and at all but one the level was said to be <10 nCi/g. One plant gave analytical results showing 11.4 nCi/g, but since they solidified their resin, the final waste form contained <10 nCi/g and was thus not TRU waste.

4. Shipment of Spent Resin

- Amounts shipped vary widely, being generally much higher for BWRs than for PWRs. The former usually reported annual shipments of many thousand ft^3/reactor and the latter only several hundred ft^3/reactor.

- The plant reporting the highest maximum loading on a spent resin (60 Ci/ft^3) shipped 15,000 ft^3 total, of which only 100 ft^3 was the high level coolant clean up resin.

- Most of the plants reporting ship dewatered resin, but six reported using solidification and one shipped in both dewatered and solid form. Several of those plants shipping large amounts solidified them.

5.2 Published Information on Radiation Damage to Resins

Experimental results from the literature on the effects of ionizing radiation on organic ion exchange resins were examined in an effort to obtain the information required to establish a maximum permissible loading for the resins disposed of by nuclear power plants. Only strong base anion resins and strong acid cation resins of the polymerization type were considered. The principal areas of interest were gas generation, loss of exchange capacity, changes in pH, physical damage to resin beads (i.e., fragmentation), changes in flow properties (i.e., agglomeration), and enhanced corrosion of metals in contact with irradiated resins. The earlier literature reviewed by Gangwer, Goldstein, and Pillay[4] covers mostly the first three areas. Recent work[5-8] in connection with the program to clean up the contaminated water at TMI-II deals with all the areas except loss of exchange capacity. From both the earlier work and the recent work, more information is available on cation than on anion resins, but for both types a broader data base would be helpful for our purpose. Recommendations for research needed to provide a suitable data base are given in Section 3.5. Information from the literature of importance to this report is summarized below.

- Changes in appearance such as fracturing of resin beads and agglomeration have not been quantitatively measured, but qualitatively they serve as indicators of resin damage. These changes and darkening of color become noticeable in the vicinity of 10^8 rad.

- Radiation damage to resins results in either loss of functional groups or breaking of bonds in the structural framework of the resin polymer.

- Removal of functional groups by radiation causes loss of exchange capacity, formation of gas (SO_2 from cation resins and nitrogen-containing gases from anion resins), and changes in pH. The latter are marked in cation resins in the H^+ form, but may be rather small in anion resins. No information could be found on effects in mixed resins.

- Both pH change and loss of exchange capacity can be measured in cation resins at radiation doses well below 10^8 rad, and often as low as 10^7 rad.

- Damage to the polymer structural framework leads to changes in the degree of cross-linkage and to generation of gases, particularly H_2, CO and CO_2, and CH_4, from both cation and anion resins.

- Although irradiation of resin leads to net formation of gaseous products, there is marked depletion of any oxygen present during the irradiation.

- Gas generation in general is greater from anion than from cation resins. H_2 seems always to be formed in the largest amount. Some

of this H_2 is a result of radiolysis of water in the resin matrix in moist resins, but in dry resins it obviously must come from the resin itself.

- Gas generation lends itself to accurate measurement and should thus be a good monitor of radiation damage. Results in the literature show some apparent conflict, but this is likely at least partly due to lack of a sufficient data base to cover all the forms of the resins for which information is required (e.g., acid/base forms, salt forms, dry, dewatered or moist, etc).

- Results of investigations reported in the literature support a nearly linear increase in gas generation with dose, with measurable amounts produced well below 10^8 rad. One recent investigation[6] found an apparent threshold for gas production from both cation and anion resin of about 5×10^7 rad, but that has not been confirmed.

5.3 Maximum Permissible Dose

NRC's draft BTP suggesting a maximum permissible loading of 10 Ci/ft^3 on spent power plant resins for disposal pointed out that such a loading of Cs-137 would deliver a total dose of about 10^8 rad to the resin. We feel that a rule on maximum loading should be given in terms of dose rather than curie level, since damage is a function of dose, but the dose will vary greatly for the same curie level depending on the RN or mixture of RNs involved. For the longer-lived RNs on the power plant spent resins, Co-60 and Cs-137, the maximum long-term doses to resin from 10 Ci/ft^3 loadings would be 6.4×10^7 rad and 1.3×10^8 rad, respectively. Maximum doses can be readily calculated from the curie loadings by a method outlined by Swyler et al.[8]

In arriving at a maximum allowable dose, the reference resin, on the basis of power plant usage, was chosen to be a 1:1 cation:anion mixture of dewatered resin in the H^+/OH^- form. Changes of appearance such as color, bead fracturing and agglomeration become noticeable at a dose of around 10^8 rad to resin in the reference state. On the basis of these qualitative effects a maximum permissible dose would then be 10^8 rad. Loss of exchange capacity and pH change are felt not to be appropriate effects on which to base a rule. Gas generation, on the other hand is considered a suitable property for this purpose. The reference resin mixture is estimated to generate about 2.7×10^{-5} moles of gas/g of resin at 10^8 rad total dose. This is a measurable amount, but when generated over a period of many years in a burial site would present no significant hazard to the general public. However, to be conservative, and to avoid higher generation rates which could be hazardous in the short-term due to hydrogen formation during handling at power plant or burial site, a maximum permissible dose has been chosen as 10^8 rad.

The following points are relevent to the usage and treatment of reactor coolant cleanup resins.

- Sr-90 appears not likely to be found on these resins to the extent of more than a few percent of the Cs-137 activity. However, since the Sr,Y-90 couple delivers a dose per curie double that of Cs-137 and four times that of Co-60, power plants should analyze their coolant cleanup resins (and any other high-level resins) for Sr-90 and report the amount with the other RNs. Further, the contribution from Sr, Y-90 should be explicity included in dose calculations.

- Short-lived RNs in the reactor coolant can build up steady state saturation levels on the coolant cleanup resins high enough to give the resin a dose approaching 10^7 rad in six months of continuous operation. This is probably less than the uncertainty with which plants will be able to estimate long term doses of 10^8 rad, so is not a determining factor in setting a maximum dose limit. However, it is an indication that the limit of 10^8 rad based on long lived isotopes is not too conservative, since the resin obtains doses from other sources.

- Finally, limited information available in the literature(5) suggests a higher maximum loading for solidified resins might be possible. Although such higher loadings might improve economics through volume reduction, more extensive research is necessary to establish a technical basis to allow such loadings. Recommendations for that research are included in Section 3.7.3.

5.4 Development of Test Procedure for Exceptions to the Maximun Dose Rule

A test procedure has been written (Section 4.4.3) based on gas generation from a dry resin. The reference resin used for estimating the amount of gas generation to be permitted in the test irradiation is a 1:1 cation: anion mixture in the H^+/OH^- form. The amount of gas estimated to be formed from such a resin mixture at a total dose of 10^8 rad was 0.3 cc of gas at STP/g of dry resin.

Measurement of changes in exchange capacity may also be suitable as the basis for a test. The test would involve only simple wet chemical methods, and irradiations could probably be carried out in air rather than in vacuum as required for the gas generation test.

Some of the data required for estimating the amount of gas to be permitted in the test, and also the maximum total dose to be permitted in resins for disposal, were not available in the literature and had to be extrapolated from results obtained with resins of another form or different water content. Information on anion resins is particularly limited. To remedy this situation, recommendations are given in Section 3.5 for additional research needed to provide an adequate data base.

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APPENDIX A

1. Blank Copy of Questionnaire of Ion Exchange Resin Usage in Nuclear Power Plants.

2. Summary of Responses of Particular Interest to this Report. (The responses obtained from some of the power plants have been supplemented by information obtained by telephone contact with plant personnel.)

SURVEY OF ION EXCHANGE RESIN USAGE IN NUCLEAR POWER PLANTS

Nuclear Waste Management Division
Brookhaven National Laboratory
Upton, New York 11973

1. Plant name
   Unit(s)/Type
   Utility Company

2. Demineralizer system:	Condensate or Boron Recycle	Reactor Coolant	Radwaste	Fuel Pool	Other
3. Type of operation: Deep bed, Powdex, etc. (Specify combinations)					
4. Type of bed: Cation, anion, mixed If mixed, stratified or homogeneous beds? % cation/% anion Bed dimensions Volume of resin in bed					
5. Resin(s) identification: Manufacturer/Vendor Tradename(s) Ionic form used in bed (e.g., H^+, OH^-, Li^+, etc.)					
6. Do you use any inorganic ion exchange resins? (Please specify)					
7. Do resins received from manufacturer/vendor contain any freestanding liquids?					

8. Characteristics of influent:
   - Flow rate (gpm)
   - Temp (°F)
   - Pressure (psig)
   - pH
   - Conductivity (μmho/cm)
   - Gross α activity (μCi/mL)
   - Gross β,γ activity (μCi/mL)
   - Principal radionuclides (μCi/mL)
     (Attach list if available)
   - Total suspended solids (ppb)
   - Contaminants:
     - Chemical species (e.g., Cl^-, silica, etc.)
     - Other materials (e.g, detergents, oil, etc.)

9. Characteristics of effluent:
   - Flow rate (gpm)
   - Temp (°F)
   - pH
   - Pressure (psig)
   - Conductivity (μmho/cm)
   - Gross α activity (μCi/mL)
   - Gross β,γ activity (μCi/mL)
   - Principal radionuclides (μCi/mL)
     (Attach list if available)
   - Total suspended solids (ppb)
   - Contaminants:
     - Chemical species (e.g., Cl^-, silica, etc.)
     - Other materials (e.g., detergents, oil, etc.)

10. Resin replacement:
    - Criteria for replacement (ΔP, conductivity, etc.)
      [Specify value(s)]
    - Dose rate at surface of bed vessel (R/hr)
    - Evidence of radiation damage
      (e.g., agglomeration, gas generation, color changes, etc.)

11. Spent resin holding tank(s):
    - Number of tanks
    - Volume
    - Physical structure (e.g., conical bottom, etc.)
    - Special features (e.g., nitrogen purging, etc.)
    - Mechanical properties (e.g., pressure limit, etc.)
    - Construction material (e.g., stainless steel, etc.)
    - Are resins from different beds mixed or stored separately?
      - If mixed, specify combinations
    - Maximum time resin stored here

12. Characteristics of water in spent resin holding tank:
    - pH
    - Conductivity (μmho/cm)
    - Gross α activity (μCi/mL)
    - Gross β, γ activity (μCi/mL)
    - Principal radionuclides (μCi/mL)
    - (Attach list if available)
    - Total suspended solids (ppb)
    - Contaminants:
      - Chemical species (e.g., Cl^-, silica, etc.)
      - Other materials (e.g., detergents, oil, etc.)

13. Dewatering information:
    - Process (e.g., centrifugation, etc.)
    - Are resins dewatered in container used for disposal?
    - Any problems?

14. Dewatered resin:
    - Gross α activity (μCi/mL)
    - Gross β, γ activity (μCi/mL)
    - Annual volume of resin for disposal

15. Container used for disposal of dewatered resins:
    - Manufacturer/Vendor
    - Dimensions
    - Volume
    - Physical structure (e.g., conical bottom, etc.)
    - Special features (e.g., underdrain, etc.)
    - Construction material (e.g., stainless steel, etc.)
    - Mechanical properties (e.g., pressure limits, etc.)
    - Volume of resin in container

16. Do you plan to use a high integrity container in the future?

17. For non-solidified resins stored on site:
    Average total volume stored
    Average storage time
    Location and conditions (indoor/outdoor, temperature range, etc.)

18. During storage on site, are containers checked periodically for changes in:
    Freestanding liquid (volume, pH, etc)?
    Gross α activity?
    Gross β,γ activity?
    Temperature?
    Pressure?
    Container integrity (e.g., leakage, corrosion, etc.)?
      (Specify any values determined.)
    Which of the above changes (or any others) are attributed to radiation damage?

19. Prior to shipment, are containers with non-solidified resins checked for changes in:
    Freestanding liquid (volume, pH, etc.)
    Gross α activity?
    Gross β,γ activity?
    Temperature?
    Pressure?
    Container integrity (e.g., leakage, corrosion, etc.)?
      (Specify any values determined.)
    Which of the above changes (or any others) are attributed to radiation damage?

20. Estimate the maximum activity (Ci/ft^3) on spent resin in a liner shipped to date.

21. Estimate the maximum activity (Ci/ft^3) on spent resin in a bed to date.

22. Do any of the resins receive radiation from an external source? (Please specify.)

23. Final disposal site(s): ☐ Barnwell, SC ☐ Beatty, NV ☐ Hanford, WA

24. Name and telephone number of person to whom questions can be addressed.

25. Additional comments on experience with ion exchange resins regarding: the drainable liquids and radiation damage to the resins.

Table A.1

Summary of Pertinent Results From Survey of Organic Ion Exchange Resin Usage by LWRs

	Type of Resin Used for Treating Reactor Coolant	Type of Operation	Activity in Coolant Water μCi/mL	Principal Radionuclides in Coolant[a]	Dose Rate at Surface of Bed Vessel[b] R/h	Criteria for Resin Replacement
BWRs						
Brown's Ferry	Mixed, 1:1 cation: anion H+,OH- form	Powdex	not answered	not answered	not available	conductivity >0.1 μmho
Cooper	Mixed cation: anion H+, OH- form	Powdex	0.1 in effluent	Co-58,60; Cr-51; Mn-54; Sr-92; Tc-99m; Ag-110m; Zn-65	not available	conductivity, ΔP, or 8-day service
Dresden	Mixed, 2:3 cation: anion	Deep Bed	1 in influent 0.1 in effluent	I-131,132,133,134,135; Ba-139,140,141; Sr-91, 92; Cs-138; Mo-99; Tc-101,104	50-200	SiO_2 > 0.3 ppm
Duane Arnold	Mixed 2:3 cation: anion	Powdex	not answered	Cr-51; Mn-54; Co-58, 60; Fe-59; Na-24	not availabe	Δp
Edwin I. Hatch	Mixed 1:1 cation: anion	Powdex	0.1 in influent 0.001-0.01 in effluent	not answered	not answered	conductivity or 7-day service
James A. Fitzpatrick	Mixed 1:1 cation: anion	Powdex	not answered	not answered	not answered	High SiO_2 conductivity
Millstone	Mixed 2:3 cation: anion	Deep Bed	0.1-1 in influent 10^{-4} in effluent	Co-60; Cs-134,137; Fe-55; Mn-54	not known	Δp > 30 psi Cl^- removal capacity
Monticello	Mixed 1:1 cation: anion H+, OH-	Powdex	not answered	not answered	not answered	2 weeks service
Peach Bottom	Mixed 1:1 cation: anion	Powdex	0.05-2 in influent	not answered	7-12 (fuel pool)	Δp = 25 psi conductivity
Vermont Yankee	Mixed	Powdex	not answered	not answered	not able to answer	conductivity
PWRs						
Beaver Valley	Mixed	Deep Bed	0.5 in influent	Co-60, Cs-137	≤500	low DF
Calvert Cliffs	Mixed 1:1 cation: anion H+, OH-	Deep Bed	not answered	not answered	not answered	
Fort Calhoun	Mixed 1:1 cation: anion	Deep Bed	1.9 in influent	I-131,132,133,134, 135; Cs-138; Co-58 Mn-54	not answered	low DF

Table A.1, Continued

Summary of Pertinent Results From Survey of Organic Ion Exchange Resin Usage by LWRs

	Type of Resin Used for Treating Reactor Coolant	Type of Operation	Activity in Coolant Water μCi/mL	Principal Radionuclides in Coolant[a]	Dose Rate at Surface of Bed Vessel[b] R/h	Criteria for Resin Replacement
Donald C. Cook	Mixed cation: anion	Deep Bed	0.2 in influent 0.02 in effluent	Cs-138; Rb-88,89; Na-24; I-131,133; Y-88; Mn-54	500	Replace at each operating cycle
Joseph M. Farley	Mixed 3:2:3 cation: anion: lithiated	Deep Bed	not answered	not answered		
Kewaunee	Mixed 1:1 cation: anion plus separate cation and anion	Deep Bed	0.1 in influent, 0.02 in effluent	Na-24, F-18	not able to measure	Low DF
Maine Yankee	Mixed 1:1 cation: anion H+, OH-	Deep Bed	1 in influent	Co-58,60; Mn-54; iodines, cesiums, and rubidiums	500-1000	DF < 1.0
Point Beach	Mixed, cation and anion	Deep Bed	0.3 in influent	I-131,132,133,134, 135; Cs-138; Rb-88; F-18; Na-24	not answered	Low DF
Prairie Island	Mixed 1:1 cation, anion Li+, OH-	Deep Bed	not answered	not answered	not answered	Low DF
St. Lucie	Mixed 2:1 cation: anion H+, OH-	Deep Bed	1.74 in influent	I-131,132,133,134, 135; Rb-88; Mo-99; Cs-138; Te-132; Y-91	not answered	Low DF
Trojan	Cation and Mixed Li+, H+, and OH-	Deep Bed	2-100	not answered	not answered	Low DF
Yankee-Rowe	Mixed 2:3 cation: anion H+, OH-	Deep Bed	1 in influent 0.01 in effluent	Cs-138; I-131,132, 133,134,135; Ba-139; Na-24; Mn-54; Se-75; Co-58; Nb-95	5-100	Δp
Zion	Mixed 1:1 cation: anion Li+, OH-	Deep Bed	0.05-0.5 in influent; 0.001 in effluent	Co-58,60; Cr-51; Cs-134,138; Ba-140; Mo-99; Zn-65; I-132	∿100	Low DF

[a]Principal RNs listed in approximate order of abundance by element. Where there is more than one isotope of the same element, they are listed together for convenience, although one or more isotopes may be out of order in terms of abundance.
[b]On bed for treating reactor coolant unless specified otherwise.

Table A.2

Summary of Pertinent Results From Survey of Organic Ion Exchange Resin Usage by LWRs

	Evidence of Radiation Damage	Activity on Dewatered Resin μCi/mL[a]	Principal Radionuclides on Dewatered Resin[a,b]	Method of Disposal
BWRs				
Brown's Ferry	none	2.5		dewatered resin
Cooper	none	0.1		solidification in concrete
Dresden	none	not known	Co-58,60; Mn-54 Cs-134,137	solidification in concrete
Duane Arnold	none	maximum 85	not answered	Hittman solidification system
Edwin I. Hatch	not answered	100-1000	Cs-134,137; Zn-65; Co-58,60	solidification
James A. Fitzpatrick	not answered			dewatered resin
Millstone	none	1 to 6		dewatered resin
Monticello	not answered			solidification in cement
Peach Bottom	none	2 to 20	Zn-65; Co-58,60 Cs-134,137; Cr-51; I-131; Mn-54	dewatered resin
Vermont Yankee	not answered	140	Cs-134,137; Zn-65; Co-58,60; Zr-95; Mn-54	dewatered resin
PWRs				
Beaver Valley	none		Co-58,60; Cs-134,137; I-131; Mn-54; Fe-59	dewatered resin
Calvert Cliffs	not answered			dewatered resin
Fort Calhoun	not answered			dewatered resin

Table A.2, Continued

Summary of Pertinent Results From Survey of Organic Ion Exchange Resin Usage by LWRs

	Evidence of Radiation Damage	Activity on Dewatered Resin μCi/mL[a]	Principal Radionuclides on Dewatered Resin[a,b]	Method of Disposal
PWRs, Continued				
Donald C. Cook	none	~50	Co-58,60; Cs-134,137 Mn-54; Sb-124	dewatered resin
Joseph M. Farley	none	1 to 100	Co-58,60; Mn-54; Cs-137; H-3	dewatered resin
Kewaunee	none			solidification
Maine Yankee	none	1000 to 5000[c]	Co-58,60; Mn-54; Cs-134,137; Sr-89,90	dewatered resin
Point Beach	none		Co-58,60; Cs-134,137 Sb-125; Ru-106; Mn-54 Cr-51	dewatered resin
Prairie Island	not answered	2 to 10		dewatered resin
St. Lucie	not answered	up to 140		dewatered resin
Trojan	not answered	35	Co-58,60; Cs-134,137 Mn-54; Ce-144	dewatered resin and solidification
Yankee-Rowe	none	typical 0.1-5 up to 50		dewatered resin
Zion	none		Co-58,60; Mn-54; Cs-134,137; I-131; Cr-51	solidification

[a]In general, on resin from reactor coolant cleanup.
[b]Principal RNs listed in approximate order of abundance by element. Where there is more than one isotope of the same element, they are listed together for convenience, although one or more isotopes may be out of order in terms of abundance.
[c]Largely Co-58 (half-life of 71 days).

Table A.3

Summary of Pertinent Results From Survey of Organic Ion Exchange Resin Usage by LWRs

	Disposal Container	Maximum Activity on Resin Shipped to Date Ci/ft^3	Maximum Activity on Spent Resin in a Bed Ci/ft^3	Resin Shipped Annually per Reactor ft^3	Other Comments
BWR's					
Brown's Ferry	carbon steel 160 ft^3	3.7	beds not used low on precoats	10,000	
Cooper	55-gal drum	2	beds not used	1,200	
Dresden	55-gal drum	18 (before solidification)	18	300	activity on resin 75% activation products
Duane Arnold	HN-600 liner, 85 ft^3	0.15	1.55	6,000	gas pressure developed from bacterial decomposition, but not from radiation damage
Edwin I. Hatch	55-gal drum	60 (before solidification)	60	15,000	ship only 100 ft^3/yr of reactor coolant cleanup resin. 90% FP on coolant cleanup resin
James A. Fitzpatrick	Chem-Nuclear 85 ft^3 high density polyethylene			1,000	
Millstone	Chem-Nuclear 195 ft^3	2.91	~3	5,000	
Monticello					
Peach Bottom	55-gal 17H Steel Drum	Maximum this year 0.3		25,000	activity on resin 80% activation products
Vermont Yankee	Brennan Weldment 170 ft^3 and 70 ft^3	~10	~10	6,200	activation products; fission products ~1:1 on resin
PWRs					
Beaver Valley	Chem-Nuclear steel 25 ft^3	~10	14.8		
Calvert Cliffs	Hittman 170 ft^3				

Table A.3, Continued

Summary of Pertinent Results From Survey of Organic Ion Exchange Resin Usage by LWRs

	Disposal Container	Maximum Activity on Resin Shipped to Date Ci/ft^3	Maximum Activity on Spent Resin in a Bed Ci/ft^3	Resin Shipped Annually per Reactor ft^3	Other Comments
Fort Calhoun	CNSI 126 ft^3	2.7	not answered	100	
Donald C. Cook	HN-100, HN-200, HN-600 liners	6.4	17	300	activity on resin 80% activation products
Joseph M. Farley	Hittman 80, 85, 170 ft^3	~3	7	300	
Kewaunee	not answered			150	
Maine Yankee	Chem-Nuclear liner 126 ft^3	~125[a]	1500 R/h (distance not specified)	200	activity on resin mostly activation products
Point Beach	Chem-Nuclear 58 & 85 ft^3	33.1	not much >33	500-1000	maximum loading was unusually high; activity on resin 90% activation products
Prairie Island	Hittman and Chem-Nuclear 80 ft^3	1	2		
St. Lucie	Chem-Nuclear 85 & 200 ft^3	~ 6	~6	~850	
Trojan	Nuclear Packaging 50 ft^3	5.4	5.4	~350	beta activity on resin 80% activiation products; alpha activity on resin sometimes >10 nCi/g, but after solidification always <10 nCi/g
Yankee-Rowe	carbon steel 80 ft^3 cylinder	3	20	80-160	activity on resin 90% fission products
Zion		30 (before solidification)	40	5-10,000	activity on resins mostly activation products

[a]Largely Co-58 (half-life of 71 days).

APPENDIX B

ANALYTICAL DATA SHEETS FOR RADIONUCLIDE CONCENTRATIONS IN REACTOR COOLANT

Table B.1

Data Sheet From Dresden Unit 3

SAMPLE SOURCE D3RXH2O Sample Time 0847 Mwt

BATCH # Sample Date 21JUL81 Mwe

STEAM FLOW CU Flow Analysis Completion Date 21JUL81

CFM ON CFM OFF VS SΔT

ISOTOPE	µCi/ml	µCi/FILT.	%	ISOTOPE	µCi/ml	µCi/FILT.	%
Na-24	4.2E-3			I-131	5.1E-3		
Cr-51	2.4E-3			I-132	1.3E-1		
Mn-54	4.4E-4			I-133	5.7E-2		
Co-58	2.6E-4			I-134	4.4E-1		
Fe-59	5.9E-4			I-135	1.4E-1		
Co-60	1.3E-3			Xe-135			
Sr-89				Cs-134			
Sr-90				Cs-136	1.5E-4		
Sr-91	3.2E-2			Cs-137	1.8E-5		
Sr-92	8.6E-2			Cs-138	6.9E-2		
Zr-95	1.5E-4			Ba-139	1.7E-1		
Nb-95m				Ba-140	6.3E-3		
Nb-95	1.6E-4			Ba-141	1.8E-1		
Mo-99	9.5E-3			La-140	6.3E-3		
Tc-99m	6.5E-2			Ce-141	2.9E-4		
Tc-101	4.6E-1			Ce-144			
Ru-103	2.7E-4			Np-239	6E-3		
Tc-104	4.2E-1			As76	3.4E-4		
Ag-110m	7.7E-6			ZN65	7.3E-5		
Sb-124	6.5E-5						

Table B.2

Data Supplied by St. Lucie
Copy of Addendum II to Table 31 of Clark's 1978 Report(2)

Addendum I : Ion exchange systems – radionuclides present

Isotope	Reactor coolant system		Boric acid recycle		Fuel pool		Waste system	
	Conc.[a]	Physical form[b]	Conc.[a]	Physical form[b]	Conc.[a]	Physical form[b]	Conc.[a]	Physical form[b]
^{3}H	1.47 (−1)	DG+L	1.47 (−1)	DG+L	1.47 (−1)	DG+L	5.29 (−3)	DG+L
^{84}Br	6.40 (−3)	DG+L	0		0		0	
^{85m}Kr	2.05 (−1)	DG	0		0		0	
^{85}Kr	1.22 (−1)	DG	6.10 (−2)	DG	6.10 (−2)	DG	0	
^{87}Kr	1.12 (−1)	DG	0		0		0	
^{88}Kr	3.59 (−1)	DG	0		0		0	
^{86}Rb	3.52 (−1)	DS	0		0		0	
^{89}Rb	8.80 (−3)	DS	0		0		0	
^{89}Sr	6.99 (−4)	DS	6.18 (−6)	DS	6.18 (−6)	DS	3.42 (−8)	DS
^{90}Sr	3.60 (−5)	DS	3.60 (−7)	DS	3.60 (−7)	DS	5.44 (−9)	DS
^{90}Y	1.41 (−4)	C	1.36 (−5)	C	1.36 (−5)	C	3.87 (−10)	C
^{91}Sr	4.91 (−4)	DS	0		0		0	
^{91}Y	1.53 (−2)	C	1.38 (−2)	C	1.38 (−2)	C	8.29 (−7)	C
^{99}Mo	2.80 (−1)	DS	3.00 (−2)	DS	3.00 (−2)	DS	8.22 (−7)	DS
^{103}Ru	5.70 (−4)	C	4.87 (−5)	C	4.87 (−5)	C	2.37 (−8)	C
^{106}Ru	3.42 (−5)	C	3.36 (−6)	C	3.36 (−6)	C	4.03 (−9)	C
^{129}Te	3.50 (−3)	DS	0				0	
^{129}I	9.95 (−9)	DS	0		0		0	
^{131}I	5.48 (−1)	DS	2.52 (−4)	DS	2.52 (−4)	DS	5.74 (−6)	DS
^{131m}Xe	2.04 (−1)	DG	6.07 (−2)	DG	6.07 (−2)	DG	0	
^{132}Te	4.55 (−2)	DS	6.92 (−4)	DS	6.92 (−4)	DS	1.71 (−7)	DS
^{132}I	1.50 (−1)	DS	0		0		0	
^{133}I	7.81 (−1)	DS	6.25 (−7)	DS	6.25 (−7)	DS	1.43 (−7)	DS
^{133}Xe	2.49 (+1)	DG	3.82 (+1)	DG	3.82 (+1)	DG	0	
^{134}Te	3.06 (−3)	DS	0		0		0	
^{134}I	8.56 (−2)	DS	0		0		0	
^{134}Cs	1.38 (+2)	DS+C	1.37 (−2)	DS+C	1.37 (−2)	DS+C	1.84 (−6)	DS+C
^{135}I	3.72 (−1)	DS	7.35 (−14)	DS	7.35 (−14)	DS	0	
^{135}Xe	1.64 (+1)	DG	0		0		0	
^{136}Cs	3.50 (−3)	DS+C	2.20 (−3)	DS+C	2.20 (−3)	DS+C	5.82 (−8)	DS+C
^{137}Cs	4.42 (−2)	DG	4.41 (−2)	DS+C	4.41 (−2)	DS+C	6.67 (−6)	DS+C
^{138}Xe	4.97 (−2)	DG	0		0		0	
^{138}Cs	9.52 (−2)	DS	0		0		0	
^{140}Ba	8.43 (−4)	DS+C	5.18 (−6)	DS+C	5.18 (−6)	DS+C	1.37 (−8)	DS+C
^{140}La	8.07 (−4)	C	1.95 (−6)	C	1.95 (−6)	C	9.42 (−10)	C
^{143}Pr	8.06 (−4)	C	5.11 (−5)	C	5.11 (−5)	C	1.39 (−8)	C
^{144}Ce	5.70 (−4)	C	5.57 (−5)	C	5.57 (−5)	C	6.31 (−8)	C
^{60}Co	7.16 (−4)	DS+C	7.14 (−5)	DS+C	7.14 (−5)	DS+C	1.03 (−7)	DS+C
^{59}Fe	2.94 (−5)	C	2.56 (−6)	C	2.56 (−6)	C	1.33 (−9)	C
^{58}Co	6.40 (−3)	DS+C	5.89 (−4)	DS+C	5.89 (−4)	DS+C	3.91 (−7)	DS+C
^{54}Mn	3.79 (−5)	DS+C	3.72 (−6)	DS+C	3.72 (−6)	DS+C	4.30 (−9)	DS+C
^{51}Cr	5.20 (−3)	DS+C	4.19 (−4)	DS+C	4.19 (−4)	DS+C	1.65 (−7)	DS+C
^{95}Zr	1.29 (−6)	C	1.17 (−7)	C	1.17 (−7)	C	0	

[a]Values listed as μCi/cc; numbers in ( ) are powers of 10. Data are expressed for 0.1% failed fuel.
[b]DG = dissolved gas; L = liquid; DS = dissolved solids; C = crud or particulates.

Table B.3

Data Sheet From Donald C. Cook Unit 1

LIST OF RADIONUCLIDES

SPENT FUEL PIT		
Cs - 134	3.590 E-4	μCi/ml
Cs - 137	1.234 E-3	μCi/ml
Co - 58	1.184 E-3	μCi/ml
Co - 60	3.975 E-3	μCi/ml
U-1 REACTOR COOLANT CRUD		
I - 131	9.061 E-5	μCi/cc
Cr - 51	8.781 E-5	μCi/cc
I - 133	7.951 E-5	μCi/cc
Cs - 134	1.297 E-5	μCi/cc
Ag - 110m	4.589 E-5	μCi/cc
Zr - 95	1.398 E-5	μCi/cc
Nb - 95	2.759 E-5	μCi/cc
Co - 58	2.466 E-4	μCi/cc
Mn - 54	1.695 E-5	μCi/cc
CO - 60	6.940 E-5	μCi/cc
U-1 REACTOR COOLANT		
I - 131	8.170 E-3	μCi/ml
Xe - 133	1.564 E-1	μCi/ml
Kr - 85m	8.013 E-3	μCi/ml
Kr - 88	5.068 E-3	μCi/ml
Xe - 133m	6.299 E-3	μCi/ml
Xe - 135	6.486 E-2	μCi/ml
Kr - 87	4.979 E-3	μCi/ml
Cs - 138	3.603 E-2	μCi/ml
I - 133	3.926 E-3	μCi/ml
Mn - 54	1.978 E-3	μCi/ml
Y - 88	3.820 E-3	μCi/ml
Rb - 88	1.527 E-2	μCi/ml
Rb - 89	3.348 E-3	μCi/ml
Ar - 41	1.895 E-3	μCi/ml
Na - 24	8.929 E-3	μCi/ml

U-1 Primary Water Storage Tank
No Activity Detected

Steam Generator Blowdown Flash Tank
No Activity Detected

Table B.4

Data Sheet From Fort Calhoun

FORT CALHOUN STATION (OPPD) 17-AUG-81 10:20:14

R A D I O N U C L I D E A N A L Y S I S R E P O R T

NUCLIDE	ACTIVITY IN UC/GM MEASURED	ERROR	DECAY CORRECTED	ERROR	ENERGY COMPARISON (KEV) EXPECT	DIF
XE-133	9.70E-01	+-6.94E-03	9.74E-01	+-6.97E-03	81.00	0.0
CO-57	MDA<5.26E-04		MDA<5.26E-04		122.06	
MO-99	MDA<4.69E-04		MDA<4.72E-04		140.51	
CE-141	7.09E-03	+-2.30E-04	7.09E-03	+-2.30E-04	145.40	0.0
SN-117M	MDA<5.21E-04		MDA<5.22E-04		158.40	
XE-135	1.96E-01	+-1.39E-03	2.07E-01	+-1.46E-03	249.60	0.2
CR-51	MDA<6.11E-03		MDA<6.11E-03		320.03	
I-131	5.12E-02	+-4.32E-04	5.13E-02	+-4.33E-04	364.46	0.1
RU-103	MDA<7.62E-04		MDA<7.62E-04		497.08	
I-133	8.07E-02	+-1.12E-03	8.25E-02	+-1.15E-03	529.90	0.0
BA-140	MDA<2.74E-03		MDA<2.75E-03		537.25	
CS-137	MDA<1.30E-03		MDA<1.30E-03		661.64	
ZR-95	MDA<1.51E-03		MDA<1.51E-03		756.72	
NB-95	4.37E-03	+-2.57E-04	4.37E-03	+-2.57E-04	765.79	0.5
CS-134	MDA<1.04E-03		MDA<1.04E-03		795.80	
CO-58	1.56E-02	+-4.18E-04	1.56E-02	+-4.18E-04	810.76	-0.0
MN-54	7.19E-03	+-3.79E-04	7.19E-03	+-3.79E-04	834.83	0.1
I-134	6.15E-02	+-8.62E-04	1.05E-01	+-1.47E-03	884.18	-0.0
					847.08	-0.0
I-132	7.52E-02	+-3.86E-03	9.26E-02	+-4.75E-03	954.60	0.1
					667.70	0.1
					772.70	0.0
CS-138	1.28E-01	+-2.13E-03	3.01E-01	+-4.98E-03	1009.70	0.0
					462.70	0.1
					1435.70	0.3
CS-136	MDA<1.22E-03		MDA<1.22E-03		1048.10	
FE-59	MDA<1.79E-03		MDA<1.80E-03		1099.22	
ZN-65	MDA<1.91E-03		MDA<1.91E-03		1115.52	
I-135	9.44E-02	+-1.95E-03	1.01E-01	+-2.10E-03	1260.50	0.0
					1131.60	0.0
CO-60	MDA<9.49E-04		MDA<9.49E-04		1332.48	
LA-140	MDA<9.74E-04		MDA<9.85E-04		1596.40	
SB-124	MDA<2.18E-03		MDA<2.18E-03		1691.00	

STANDARD DEVIATION = 0.1

ERROR QUOTATION AT 1.00 SIGMA
MDA CONFIDENCE LEVEL AT 90.0%

Table B.5

Data Sheet From Point Beach Unit 1

UNIT 1 REACTOR COOLANT EBAR CALCULATION

ALL AVERAGE AND TOTAL ENERGIES LISTED AS MEV :

ISOTOPE	BETA	GAMMA	CE	EI	AI (UCI/CC)	EIAI
I-131	0.1970	0.3710	0.0000	0.5680	3.10000000E-02	0.0176
I-132	0.4480	2.4000	0.0000	2.8480	2.11000000E-01	0.6009
I-133	0.4230	0.4770	0.0000	0.9000	2.01000000E-01	0.1809
I-134	0.4550	1.9400	0.0000	2.3950	4.54000000E-01	1.0873
I-135	0.3080	1.7800	0.0000	2.0880	2.66000000E-01	0.5554
XE-133	0.1150	0.0300	0.0280	0.1730	1.34000000E-01	0.0232
KR85M	0.2330	0.1510	0.0000	0.3840	9.22000000E-02	0.0354
XE133M	0.0000	0.0325	0.2100	0.2425	1.71000000E-04	0.0000
KR-88	0.3410	1.7400	0.0000	2.0810	1.52000000E-01	0.3163
XE-135	0.3220	0.2460	0.0000	0.5680	3.29000000E-01	0.1869
KR-87	1.0500	1.3700	0.0000	2.4200	5.26000000E-02	0.1273
AR-41	0.4040	1.2770	0.0000	1.6810	8.83000000E-03	0.0148
KR-85	0.2230	0.0021	0.0000	0.2251	6.09000000E-03	0.0014
XE135M	0.0000	0.4220	0.0974	0.5194	9.26000000E-02	0.0481
X 138	0.8000	2.8700	0.0000	3.6700	1.37000000E-01	0.5028
F-18	0.2120	0.9910	0.0000	1.2030	6.94000000E-02	0.0835
MO-99	0.3800	0.1370	0.0000	0.5170	1.96000000E-03	0.0010
CS-134	0.1660	1.5900	0.0000	1.7560	1.36000000E-03	0.0024
CS-137	0.1730	0.5630	0.0730	0.8090	2.81000000E-03	0.0023
CS-138	1.1300	2.0800	0.0000	3.2100	2.78000000E-01	0.8924
CO-58	0.0237	0.9770	0.0000	1.0007	9.58000000E-05	0.0001
CO-60	0.1050	2.5100	0.0000	2.6150	2.13000000E-05	0.0001
NA-24	0.4630	4.1230	0.0000	4.5860	7.49000000E-03	0.0343
MN-54	0.0002	0.8350	0.0000	0.8352	3.10000000E-06	0.0000
TE-132	0.1000	0.2160	0.0477	0.3637	8.74000000E-04	0.0003
FE-59	0.1260	1.1900	0.0000	1.3160	3.04000000E-06	0.0000
LA-140	0.3970	2.1200	0.0000	2.5170	1.58000000E-04	0.0004
CS-136	0.1390	2.2300	0.0000	2.3690	1.16000000E-04	0.0003
RB-88	1.6100	0.5700	0.0000	2.1800	2.17000000E-01	0.4731

TOTAL GAS CONCENTRATION (UCI/CC) : 1.00449100E+00
TOTAL MCA LIQUID CONCENTRATION (UCI/CC) : 1.74229124E+00
ICN 15 MINUTE DECAY (UCI/CC) : 2.0791
RATIO MCA LIQ. ACT./ICN ACT. : 0.8380

TOTAL EBAR: (SUM EIAI)/(GAS.ACT.+ MCA LIQ.ACT. EXC. IODINES) = 1.7340

TECH SPEC COOLANT ACTIVITY LIMIT: (100/EBAR) (CI/M3) = 57.6696

PERCENT OF TECH SPEC ALLOWANCE PRESENTLY (INCL. IODINES): 4.7630

APPENDIX C

ANALYTICAL DATA SHEETS FOR RADIONUCLIDE LOADINGS ON SPENT RESIN FROM REACTOR COOLANT CLEANUP

Table C.1

Data Sheet From Donald C. Cook Unit 1

```
IS WEIGHTED DECAY TIME TO BE USED? NO
SAMPLE DATE    21 6 81
IS 21 JUN, 1981 CORREC. ? YES
SAMPLE TIME    0928
DATE ANALYZED    21 6.81
IS 21 JUN, 1981 CORRECT ? YES
TIME ANALYZED    1123
DECAY TIME:      115.00 MIN
REGION:  1
GEOMETRY:  10
KEV WINDOW:  2
# OF SIGMA UNITS:  2
# OF SMOOTHS:  1
DEAD TIME (%):  5
ANALYZ MULTIPLETS?
SAMPLE ID+CONFIG: SPENT RESIN TANK #6453-R
SAMPLED BY: BS
ANALYZED BY:  ERS
RX POWER(RCS SAMPLES ONLY):  NA

.......................................................
D.C.COOK PLANT
SAMPLE ID:  SPENT RESIN TANK #6453-R
SAMPLED BY:  BS
ANALYZED BY:  ERS
RX POWER:  NA
.......................................................

COLLECT TIME:        1000.  (SEC)

.......................................................
                RADIONUCLIDE ANALYSIS

ISOTOPE                ACTIVITY

CO-57          7.03033E-04 UC/ML +- 25.44 %
I-131          3.43120E-03 UC/ML +- 12.07 %
CS-134         3.87059E-03 UC/ML +-  7.14 %
SB-124         8.87810E-03 UC/ML +-  7.14 %
CS-137         1.44026E-02 UC/ML +-  4.24 %
NB-95          1.04387E-03 UC/ML +- 36.45 %
CO-58          1.54701E-01 UC/ML +-  0.69 %
MN-54          8.02582E-03 UC/ML +-  5.38 %
CO-60          5.47894E-02 UC/ML +-  1.47 %
```

Table C.2

Loading Calculation From Donald C. Cook Unit 1

DEMINERALIZER RESIN CALCULATION SHEET

Isotope	Concentration from Lab	(a) Fraction of Total Concentration	(b) τ/10	(c) Exposure Contribution (a) x (b)	Total Activity in Resin (a) x (e)
MN-54	8.026×10^{-3}	.0332	0.47	.0156	13.766
CO-57	7.030×10^{-4}	.0029	0.09	.000261	1.202
CO-58	1.547×10^{-1}	.6396	0.55	.35178	265.206
CO-60	5.479×10^{-2}	.2265	1.32	.29898	93.917
Ag-110m			1.43		
I-131	3.431×10^{-3}	.0142	0.22	.003124	5.8880
CS-134	8.871×10^{-3}	.0367	0.87	.031929	15.217
CS-137	1.440×10^{-3}	.0059	0.33	.001947	2.446
La-140			2.37		
Zr-95			0.41		
Nb-95	1.044×10^{-3}	.0043	0.42	.001806	1.783
Sb-124	8.878×10^{-3}	.0367	0.98	.035966	15.217
TOTAL	2.4188×10^{-1}	1.00		.741393	414.643

Exposure Contribution (c) = Fraction of total concentration (a) x τ/10 (b).

Exposure at 1 meter from 100 ml sample = .0167 R/hr.

if reading taken at 3 ft., R/hr at 1 meter = R/hr at 3 ft. x .836

.020 x .836

(B) Resin volume factor = ft^3 in cask x 283.2

= 65 x 283.2

= 18408 (d)

(A) Total activity in resin = $\frac{\text{Exposure at 1 meter x resin volume factor (d)}}{\text{Total exposure contribution (c)}}$

$= \frac{(.0167 \text{ R/hr} \times 18408)}{(.741393)}$

= 414.6433 Ci(e)

Performed by [signature]

Approved by [signature]

Table C.3

Data Sheet From E. I. Hatch Unit 1

Summary of Analysis

Unit I
CUPS Resin

CONCENTRATIONS

ISOTOPE	ENERGY (Kev)	NET ACTIVITY (uCi/g)	ERROR (uCi/g)	L L D (uCi/g)	% Abundance	FLAG
Selected Nuclides						
Xe-133	80.99	---	9.819E-01	1.156E+00		L L D
Ce-146	98.30	---	6.523E+03	7.984E+03		L L D
Te-131m	102.20	---	4.671E+00	5.719E+00		L L D
Np-238	119.80	---	7.482E+01	9.159E+01		L L D
Ce-144	133.50	---	3.570E+00	4.376E+00		L L D
MoTc-99m	140.50	---	4.681E-01	5.696E-01		L L D
Ce-141	145.40	---	8.368E-01	1.034E+00		L L D
Kr-85M	151.00	---	1.078E+00	1.314E+00		L L D
Cs-136	17[illegible].70	---	3.532E+00	4.308E+00		L L D
Kr-88	1[illegible]5.10	---	4.165E+00	5.095E+00		L L D
Te-1[illegible]	[illegible]8.20	---	7.713E-01	9.451E-01		L L D
Xe-13[illegible]M	233.18	---	6.755E+00	8.289E+00		L L D
Xe-135	249.60	---	9.454E-01	1.149E+00		L L D
Te-134	278.10	---	2.086E+02	2.569E+02		L L D
Cr-51	320.10	---	7.101E+00	8.687E+00		L L D
La-140	328.80	---	3.657E+00	4.490E+00		L L D
I-131	364.50	---	9.596E-01	1.180E+00		L L D
Sn-113	391.71	---	1.302E+00	1.596E+00		L L D
Zr-88	392.80	---	8.609E-01	1.060E+00		L L D
Kr-87	402.70	---	1.715E+01	2.098E+01		L L D
Sb-125	427.90	---	2.918E+00	3.576E+00		L L D
Zn-69M	438.70	---	1.121E+00	1.372E+00		L L D
Cs-138	462.50	---	6.315E+02	7.749E+02		L L D
W-187	479.50	---	3.629E+00	4.430E+00		L L D
Y-87	484.80	---	9.036E-01	1.114E+00		L L D
I-133	529.90	---	1.007E+00	1.233E+00		L L D
Ba-140	537.40	---	3.292E+00	4.019E+00		L L D
Cs-134	603.87	5.465E+02	3.057E+00	2.235E+00	37.38	Measured
Cs-137	660.75	7.571E+02	3.667E+00	2.344E+00	51.78	Measured
Ce-144	696.50	---	3.931E+01	4.760E+01		L L D
Zr-95	756.90	---	1.021E+00	1.265E+00		L L D
Nb-95	765.80	---	5.787E-01	7.171E-01		L L D
Co-58	810.60	---	6.472E-01	7.956E-01		L L D
Mn-54	834.80	---	5.846E-01	7.618E-01	[illegible]	L L D
Fe-59	1099.30	---	7.469E-01	9.222E-01		L L D
Zn-65	1114.52	1.284E+02	2.954E+00	2.360E+00	8.78	Measured
Na-22	1274.55	---	2.841E-01	3.574E-01		L L D
Ar-41	1293.60	---	1.394E+00	1.804E+00		L L D
Co-60	1331.57	3.012E+01	9.911E-01	7.166E-01	2.06	Measured
Cu-64	1345.90	---	7.039E+01	8.520E+01		L L D
Na-24	1368.55	---	6.146E-01	6.878E-01		L L D
Sr-92	1386.00	---	8.577E-01	1.108E+00		L L D
Ni-65	1481.70	---	2.392E+00	3.250E+00		L L D
K-42	1524.70	---	1.261E+00	1.492E+00		L L D
BaLa-140	1596.20	---	1.820E-01	2.135E-01		L L D
Tc-104	1676.70	---	1.900E+03	1.791E+03		L L D
Sb-124	1691.00	---	2.592E-01	3.127E-01		L L D
Mn-56	1811.00	---	8.759E-01	8.415E-01		L L D

TOTAL μCi/gRAM = 1462.12

Table C.4

Data Sheet From Maine Yankee

MAINE YANKEE ATOMIC POWER COMPANY
RADIOCHEM LAB RESULTS

CONFIGURATION: 0.0100 g RESIN
SAMPLED BY: ?
DATE: 3/11/80 TIME: ? MARCH 80
ANALYZED BY: LACH
DATE: 3/11/80 TIME: 1700

GAMMA SPECTROGRAPHIC ANALYSIS

CHANNEL	AREA	ENERGY	ISOTOPES
91.67	895.	47.54	
166.01	350.	84.57	
1022.35	3497.	511.28	~~F-18~~, CO-58
1210.55	961.	605.09	CS-134
1324.84	1298.	662.06	CS-137
1594.25	578.	796.39	CS-134, ~~BI-214~~
1624.03	10075.	811.24	CO-58, ~~I-132~~
2251.22	390.	1173.94	CO-60, ~~NP-87~~
2670.35	362.	1333.17	CO-60, ~~FE-153~~
2927.73	183.	1461.61	K-40

RADIONUCLIDE ANALYSIS

ISOTOPE	ACTIVITY	
~~F-18~~	~~2.04E-01 UC/ +- 3.60 %~~	
CO-58	1.70E+00 UC/ +- 1.10 %	PER .01gm
CS-134	1.11E-01 UC/ +- 7.03 %	
CS-137	2.11E-01 UC/ +- 4.47 %	
CO-60	9.76E-02 UC/ +- 6.79 %	

ISOTOPE	THEORETICAL	MEASURED	DIFFERENCE
F-18	511.00	511.28	0.28
CO-58	810.60	811.24	0.64
CS-134	795.81	796.39	0.58
CS-137	661.64	662.06	0.42
CO-60	1332.50	1333.17	0.67

STANDARD DEVIATION= 0.54

RESULTS REVIEWED BY .....

Table C.5

Data Sheet From Peach Bottom

```
*****************************************************************
*****************      14-SEP-81 01:56:41      ****************
*****************************************************************

3B 0 SEP DRUM #2483-81   SAMPLE RESIN SCAN                    DGC

SAMPLE DATE:  14-SEP-81 00:45:00
SAMPLE IDENTIFICATION:  RESIN SAMPLE
TYPE OF SAMPLE:  BATCH ANALYSIS
SAMPLE QUANTITY:  0.5441000E-01     UNITS:  GRAMS
SAMPLE GEOMETRY:  SEP. CARD
EFFICIENCY FILE NAME:  EFF.DEC3..
*****************************************************************
                                    *
ACQUIRE DATE: 14-SEP-81 01:45:01    * FWHM(1332)          2.234
PRESET TIME(LIVE):      600. SEC    * SENSITIVITY:        5.000
ELAPSED REAL TIME:      605. SEC    * SHAPE PARAMETER : 30.0 %
ELAPSED LIVE TIME:      600. SEC    * NBR ITERATIONS:     8.
                                    *
*****************************************************************
                                    *
DETECTOR: GELI-3                    * LIBRARY:NUCL.SHP
CALIB DATE:  09-SEP-81 19:37:11     * ENERGY TOLERANCE:  2.200KV
KEV/CHNL:       0.8700811           * HALF LIFE RATIO:   8.00
OFFSET:        10.7741251 KEV       * ABUNDANCE LIMIT:  75.00%
                                    *
*****************************************************************

> RUN PB.REPORT

IDENTIFIED PEAK REPORT:
```

NUCLIDE	ENERGY KEV	NET AREA CPM	BKGD CPM	% ERR	CORRECTED UC/UNIT	CORRECTED UC/SMPL
CR-51	320.37	3.70E 01	1.32E 02	14.8	2.060E-01	1.121E-02
MN-54	835.10	7.25E 00	6.94E 01	52.7	1.097E-02	5.971E-04
CO-58	810.90	7.17E 01	8.28E 01	6.8	1.052E-01	5.726E-03
CO-60	1173.39	3.15E 02	2.85E 01	1.9	6.644E-01	3.615E-02
	1332.85	2.76E 02	9.00E 00	2.0	6.658E-01	3.623E-02
ZN-65	1115.67	6.10E 02	6.90E 01	1.4	2.454E 00	1.335E-01
I-131	364.66	1.65E 02	1.56E 02	4.2	1.267E-01	6.894E-03
CS-134	604.78	2.86E 02	1.48E 02	2.7	2.859E-01	1.556E-02
	795.94	1.65E 02	1.65E 02	4.3	2.571E-01	1.399E-02
	802.27	7.73E 00	7.18E 01	50.3	1.198E-01	6.517E-03
	1365.61	5.15E 00	5.20E 00	24.2	3.710E-01	2.019E-02
CS-137	661.52	2.92E 02	1.18E 02	2.5	4.062E-01	2.210E-02
LA-140	487.10	1.16E 01	9.00E 01	37.7	2.156E-02	1.173E-03
	1596.95	8.54E 00	2.85E 00	14.0	2.599E-02	1.414E-03

Table C.6

Data Sheet for Resin Shipment, Peach Bottom

RESIN SHIPMENT REPORT SUMMARY:

	NUCLIDE	UC/GRAM	MC/DRUM	TRANSPORT GROUP	TYPE B %	LSA %	HEAT FLUX WAT/DM
	CR-51	2.060E-01	4.021E 01	4	0.02	0.07	6.91E-06
	MN-54	1.097E-02	2.142E 00	4	0.00	0.00	1.05E-05
	CO-58	1.052E-01	2.055E 01	4	0.01	0.04	1.05E-04
	CO-60	6.644E-01	1.297E 02	3	0.06	0.22	1.99E-03
	ZN-65	2.454E 00	4.790E 02	4	0.24	0.82	1.64E-03
	I-131	1.267E-01	2.473E 01	3	0.01	0.04	8.06E-05
	CS-134	2.859E-01	5.581E 01	3	0.03	0.10	5.60E-04
	CS-137	4.062E-01	7.931E 01	3	0.04	0.14	3.61E-04
	LA-140	2.599E-02	5.075E 00	4	0.00	0.01	8.33E-05
(EST)	SR-90	4.062E-03	7.9E-01	2	0.00	0.08	9.21E-07
(EST)	FE-55	7.640E-01	1.5E 02	4	0.07	0.25	0.00E-01
(EST)	NI-63	5.979E-01	1.2E 02	4	0.06	0.20	1.18E-04

TRANSPORT GROUP	TOTAL UC/GRAM	TOTAL MC/DRUM	% TYPE A LIMIT	% TYPE B LIMIT	% LSA
1	0.000E-01	0.0E-01	0.00	0.00	0.00
2	4.062E-03	7.9E-01	1.59	0.00	0.08
3	1.483E 00	2.9E 02	9.65	0.14	0.49
4	4.164E 00	8.1E 02	4.06	0.41	1.39
5	0.000E-01	0.0E-01	0.0	0.0	NL
6	0.000E-01	0.0E-01	0.0	0.0	NL
1-6	5.651E 00	1.1E 03	15.3	0.6	2.0

MAXIMUM NUMBER OF DRUMS ALLOWED: 6 180 NL

NOTE:
NL - NO LIMIT EXISTS
NS - NO SHIPMENT ALLOWED, EXCEEDS LIMITS
***** - NUMBER TO LARGE TO PRINT

SHIPPING INFORMATION

MOST LIMITING ISOTOPE	MOST LIMITING TRANSPORT GROUP	TOTAL MC/DRUM	SHIP AS
ZN-65	4	1.1E 03	LSA

THERMAL WATTS = 4.95E-03 —

Table C.7

Data Sheet From Trojan

June 12, 1981
P. O. #N19425

GAMMA EMITTING NUCLIDES

The xxxx number indicates the sample number we have designated that particular sample for our records. The results of the gamma activity on the two filters (9363F) are expressed in terms of total microcuries. The results of the gamma activity on the resin (9363R) are expressed in terms of microcuries per gram. The weight of the resin is 2.10E-01 grams. Results are decay corrected to 5/14/81 at 1200 hours.

SAI #	9363F	9363R
Nuclide	μCi/total	μCi/gram
Cr-51	<2.22(-3)	<1.21(-1)
Mn-54	4.64±.16(-2)	2.78±.10(0)
Fe-59	<1.37(-3)	<5.87(-2)
Co-57	1.66±.29(-3)	6.07±2.4(-2)
Co-58	2.38±.13(-2)	5.32±.87(-1)
Co-60	6.04±.06(-1)	4.57±.09(1)
Zn-65	7.44±2.50(-3)	<6.71(-2)
Nb-95	5.15±.92(-3)	<2.26(-2)
Ru-106	<5.00(-3)	<1.95(-1)
Ag-110m	3.38±.88(-3)	<4.04(-2)
Sb-124	<7.52(-4)	<1.78(-2)
Sb-125	2.08±.16(-2)	<1.65(-1)
Cs-134	2.52±.09(-2)	5.13±.17(0)
Cs-137	6.76±.18(-2)	1.26±.01(1)
Ba-140	9.80±3.7(-4)	--
Ce-141	<1.02(-4)	<1.32(-2)
Ce-144	3.17±.20(-2)	3.49±1.9(-1)

Table C.8

Data Sheet From Vermont Yankee

1. Total Volume of Resin shipped (1980) - 6200 cu ft.
2. Isotopic from Low Level shipment (Condensate Demins/Radwaste)

Cs-134	7.02E-1 μCi/gm
Cs-137	2.27E+0 μCi/gm
Co-58	7.40E-2 μCi/gm
Mn-54	1.28E-1 μCi/gm
Zn-65	4.61E-1 μCi/gm
Co-60	9.24E-1 μCi/gm

Total Activity - 8780 mCi

3. Isotopic from High level shipment (Reactor Cleanups)

Tc-99m	4.19E-1 μCi/gm
I-131	5.35E-1 μCi/gm
Cs-134	1.78E+1 μCi/gm
Cs-137	4.84E+1 μCi/gm
Zr-95	1.11E+1 μCi/gm
Co-58	5.88E+0 μCi/gm
Mn-54	5.98E+0 μCi/gm
Zn-65	2.03E+1 μCi/gm
Co-60	3.05E+1 μCi/gm

Total Activity - 127.6 Curies

APPENDIX D

ANALYTICAL DATA SHEET FOR LOADING OF TRANSURANIC NUCLIDES ON SPENT REACTOR COOLANT CLEANUP RESIN

Table D.1

Data Sheet From Trojan

June 12, 1981
P. O. #N19425

ALPHA NUCLIDES

Results of 9363F are expressed in terms of microcuries per sample. Results of 9363R are expressed in terms of microcuries per gram. Activity is decay corrected to 4-01-81 at 1200 hours.

	Results	
SAI #	9363F	9363R
Nuclides	Conc. (μCi total)	Conc. (μCi/gram)
$^{239,240}Pu$	1.31±.09(-3)	1.14±.06(-2)
^{238}Pu	5.84±.41(-4)	2.96±.15(-3)
^{241}Am	1.51±.21(-5)	5.97±.99(-5)
$^{242,243}Cm$	5.12±.44(-5)	7.32±1.2(-5)
^{244}Cm	3.94±1.07(-6)	5.77±3.08(-6)

APPENDIX E

COMPUTER PROGRAM FOR CALCULATING DOSE DELIVERED TO ORGANIC ION EXCHANGE RESIN BEDS BY DIFFERENT RADIONUCLIDE LOADINGS

The program was written for interactive use in INTERCOM subsystem of the CDC 6600-15 computer at Brookhaven National Laboratory.

```
      PROGRAM DSE(INPUT,OUTPUT)
      DIMENSION CI(10),AMB(10),DDG(10),DIG(10),DCI(10),NB(10),
     1TOPE(10),HL(10),GC(10),DDB(10,10),DIB(10,10),AB(10,10)
     2,ABE(10,10),DOSE(100)
C       ENTER CONTAINER INFORMATION
      WRITE 2
      WRITE 1
    1 FORMAT(1X,"ENTER CONTAINER HEIGHT AND WIDTH IN FEET AND "
     1,"GEOMETRIC FACTOR",/,1X,3("[          ]"),/)
    2 FORMAT(1X,"PROGRAM TO CALCULATE DOSE TO RESINS IN A"
     1,/,1X," CYLINDRICAL CONTAINER BASED UPON TREATMENT IN "
     2,/,1X,"SWYLER ET AL.,BNL-NUREG-28682",//)
      READ 3,CH,CW,GB
    3 FORMAT(3G10.4)
C       ENTER ISOTOPE INFORMATION
      WRITE 5
    5 FORMAT(1X,"ENTER NUMBER OF ISOTOPES (I2):")
    7 FORMAT(I2)
      READ 7,NI
      DO 20 I=1,NI
      WRITE 9
    9 FORMAT(1X,"ENTER ISOTOPE,HALF-LIFE,NO. OF BETAS, ",/,1X,
     1"GAMMA RAY CONSTANT, AND NO. OF CURIES",/,1X,
     2"[     ][          ][][          ][          ]",/)
      READ 11,TOPE(I),HL(I),NB(I),GC(I),CI(I)
   11 FORMAT(A6,G10.4,I2,2G10.4)
      NBA=NB(I)
      AMB(I)=ALOG(2.0)/HL(I)
      WRITE 13
   13 FORMAT(1X,"ENTER AV. BETA ENERGY AND FREQUENCY ",/,1X,
     12("[          ]"),/)
      DO 20 II=1,NBA
      READ 15,ABE(I,II),AB(I,II)
   15 FORMAT(2G10.4)
   20 CONTINUE
```

APPENDIX E, Continued

COMPUTER PROGRAM FOR CALCULATING DOSE DELIVERED TO ORGANIC ION EXCHANGE RESIN BEDS BY DIFFERENT RADIONUCLIDE LOADINGS

```
C       CALCULATE CONTAINER CONSTANTS
      R=CW/2.0
      VCF=3.1416*R*R*CH
      VCC=VCF*28316.8
C       BETA DOSE CALCULATION
      DO 200 J=1,NI
      NBB=NB(J)
      DO 100 K=1,NBB
      DDB(J,K)=(CI(J)*1000./VCC)*AB(J,K)*ABE(J,K)*2.1E03
      DIB(J,K)=DDB(J,K)*8.76E03/AMB(J)
  100 CONTINUE
C       GAMMA DOSE CALCULATION
      DDG(J)=(CI(J)*1000./VCC)*GC(J)*GB
      DIG(J)=DDG(J)*8.76E03/AMB(J)
      DCI(J)=0.0
  200 CONTINUE
C       CUMULATIVE DOSE FOR THE ITH NUCLIDE
      DO 400 L=1,NI
      NBC=NB(L)
      DO 300 M=1,NBC
      DCI(L)=DCI(L)+DIB(L,M)
  300 CONTINUE
      DCI(L)=DCI(L)+DIG(L)
  400 CONTINUE
C       CALCULATE TOTAL DOSE AT TIME=T
      WRITE 420
  420 FORMAT(1X,3X,"TIME",11X,"DOSE",/,1X,3X,"(YR)",10X,"(RAD)",/)
      DO 500 N=1,33
      IF(N.EQ.1) T=1.
      IF(N.EQ.2) T=3.
      IF(N.EQ.3) T=5.
      IF(N.EQ.4) T=10.
      IF(N.GT.4) T=T+10.
      DOSE(N)=0.0
      DO 450 NN=1,NI
      EF=AMB(NN)*T
      IF(EF.GE.600.0) DOSE(N)=DOSE(N)+DCI(NN)
      IF(EF.LT.600.0) DOSE(N)=DOSE(N)+(DCI(NN)*(1-EXP(-1.0*AMB(NN)*T)))
  450 CONTINUE
      WRITE 470,T,DOSE(N)
  470 FORMAT(1X,2X,F6.2,8X,G10.4)
  500 CONTINUE
      STOP
      END
```

APPENDIX F

DETERMINATION OF DOSE DELIVERED BY SHORT-LIVED RADIONUCLIDES TO REACTOR COOLANT CLEANUP RESIN AT DRESDEN NUCLEAR POWER STATION, UNIT 3

The dose delivered by the beta and gamma emissions of a representative short-lived radionuclide, Ba-140, to the resin in the reactor coolant cleanup bed at Dresden, is calculated below. The calculations are the same for other short-lived isotopes except that the nuclear constants for each specific isotope must be used in calculating its delivered dose.

The treatment used here is based on the buildup on the resin bed of steady-state saturation levels of the isotopes being removed from the coolant stream. These levels are reached (to within 1%) in five mean-lives, and from that time on, their rates of decay are equal to their rates of buildup on the resin, assuming constant coolant flow rate through the bed. Thus, they deliver a continuous dose (and at constant dose rate as long as the reactor power is constant) to the resin all the time the bed is in service. After the bed is removed from service and the RNs decay, the dose from their decay will exactly compensate for the deficit in dose during buildup, before saturation is reached. Hence, the total dose delivered by the short-lived isotopes is simply the dose rate at saturation multiplied by the time the bed is in service.

The curie amount, C_i, of an isotope on the resin bed at its steady-state saturation value is given by

$$C_i = A_i \, F \, e_r \tau_i$$

where A_i is the activity level of the ith isotope in Ci/mL;
F is coolant flow rate through the bed in mL/min;
e_r is overall efficiency of activity removal by the bed;
τ_i is the mean-life of the ith isotope in minutes.

For the Dresden coolant cleanup system, $e_r = 0.9$ (using the reported overall decontamination factor of 10) and F is 2.27×10^6 mL/min. Taking Ba-140 as an example of a specific isotope removed from the coolant, C_i can be obtained using the mean life of Ba-140 of 2.86×10^4 min and its concentration in the coolant of 6.3×10^{-9} Ci/mL.

$$C_{Ba-140} = 370 \text{ Ci}$$

For β-decay, >99% of the β-particle energy is absorbed by the bed. Thus, the dose rate to the resin, R_i, in rad/h from β-decay of the ith isotope is obtained from the equation

$$R_i^\beta = \frac{C_i \; D \; E_i^\beta}{100 m_B}$$

where C_i is the number of curies of the ith isotope on the bed
D_β is disintegrations/h/Ci = 1.33×10^{14} d/h;
E_i^β is the average β-energy of the ith isotope in ergs;
m_B is the mass in g of the resin bed, including water. This is 5.0×10^6 g, assuming an overall density of 1 g/mL.

For Ba-140, C_i = 370 Ci, and $E_i = 6.4 \times 10^{-7}$ erg, so that R_{Ba-140} = 63 rad/h, and in a six-month operating period (plus time for complete decay of Ba-140) the total dose from β-particles delivered by Ba-140 will be 2.7×10^5 rad.

This result is the same as that obtained using a treatment based on the method of Swyler et al.[9] for calculation of absorbed β-dose due to complete radioactive decay of an isotope. This treatment uses the fact that a specific amount of an isotope trapped by the resin in unit time will deliver, during its decay, an absorbed dose calculated by the method of Swyler et al. The total β-dose delivered during the operating period of the resin bed will then be just this amount multiplied by the total time the bed is kept in service.

For β-dose determination, which is straightforward with respect to energy absorption (essentially all the β-particle energy is absorbed in the resin bed), either treatment may be used. The same is true for determination of γ-dose, but the latter requires estimation of the extent of γ-ray absorption in the resin bed. Since absorption coefficients and geometry factor are already included in the calculation of Swyler et al., it is convenient to use that method for determination of the γ-dose, keeping in mind that the calculation gives the maximum dose, due to assumption of uniform loading.

Using the notation of Swyler et al, the total γ-dose delivered by an isotope in six months, D_i^γ(total), is given by

$$D_i^\gamma(\text{total}) = D_i^\gamma(\infty) \times 2.63 \times 10^5 \text{ minutes in six months}$$

and $$D_i^\gamma(\infty) = \frac{C_i \Gamma_i \bar{g} \times 8.76 \times 10^3 \text{ h} \cdot \text{yr}^{-1}}{\lambda_i}$$

where C_i is the activity density of the ith isotope in mCi/cm^3;

Γ_i is the gamma constant for the ith isotope;

$\overline{g}$ is a geometric factor

λ_i is the decay constant of the ith isotope in yr^{-1}.

For the dimensions of Dresden's coolant cleanup bed $\overline{g}$ = 270 cm. For Ba-140, $C_i = 2.6 \times 10^{-6}$ mCi/cm^3, $\Gamma_i = 12.4$ and $\gamma_i = 19.8$ yr^{-1}. Thus, the γ-dose delivered by Ba-140 in six months is calculated to be 10.1×10^5 rad, and the total dose, both β and γ is 12.8×10^5 rad, or $\sim 1.3 \times 10^6$ rad.

NRC FORM 335 (11-81)

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

1. REPORT NUMBER (Assigned by DDC)
NUREG/CR-2830
BNL-NUREG-51565

4. TITLE AND SUBTITLE (Add Volume No., if appropriate)
Permissible Radionuclide Loading for Organic Ion Exchange Resins from Nuclear Power Plants

2. (Leave blank)

3. RECIPIENT'S ACCESSION NO.

7. AUTHOR(S)
D. R. MacKenzie, M. Lin, R.E. Barletta

5. DATE REPORT COMPLETED
MONTH June YEAR 1982

9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)
Brookhaven National Laboratory
Upton, New York 11973

DATE REPORT ISSUED
MONTH October YEAR 1983

6. (Leave blank)

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12. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)
Division of Waste Management
Office of Nuclear Material Safety and Safeguards
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555

10. PROJECT/TASK/WORK UNIT NO.

11. FIN NO.
A3159

13. TYPE OF REPORT
Technical Report

PERIOD COVERED (Inclusive dates)

15. SUPPLEMENTARY NOTES

14. (Leave blank)

16. ABSTRACT (200 words or less) A questionnaire on the use of ion exchange resins in nuclear power plants was sent to all operating reactors in the U.S. Responses were received from 23 of the 48 utilities approached. Information was sought concerning the amounts of radionuclides held by the resins, and the effects of its radiation on the resins both during operation and after removal from service. Relevant information from the questionnaires is summarized and discussed. Available literature on the effects of ionizing radiation on organic ion exchange resins has been reviewed. On the basis of published data on damage to resins by radiation, the technical rationale is given to support NRC's draft branch technical position on a maximum permissible radionuclide loading. It is considered advisable to formulate the rule in terms of a delivered dose rather than a curie loading. A maximum permissible dose of 10^8 rad is chosen because, while it is large enough that a measurable amount of damage will be done to the resin, it is small enough that the damage will be negligible at a power plant or disposal site. A test procedure has been written which a generator could use to qualify a specific resin for service at a higher dose than permitted by the general rule.

17. KEY WORDS AND DOCUMENT ANALYSIS

17a. DESCRIPTORS

1) Permissible Radionuclide Loading
2) Damage to resins by radiation
3) Low-Level Waste resin Disposal Requirements

17b. IDENTIFIERS/OPEN-ENDED TERMS

18. AVAILABILITY STATEMENT
Unlimited

19. SECURITY CLASS (This report)
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