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ENLARGED NORDIC COOPERATIVE PROGRAM ON NUCLEAR SAFETY

NUCLIDE CONTENT IN REACTOR WASTE

A Joint Scandinavian Research Project Sponsored by

The Nordic Council of Ministers

NOTE

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The views and results contained in the report are not necessarily those of the institutions who participated in the study.

Working documents belonging to the study (NKA/AOdocuments) are available upon request in each of the four participating countries, see list of addresses at the end of this report.

The results described in the present report have served as background information for volumes 1-3 of the Nordic Study on Reactor Waste, NKA/AO (81) 5.



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ABSTRACT

Certain corrosion and fission products of importance in reactor waste management cannot be measured by gammaspectrometric techniques. In this study, a method is suggested by which the occurrence of such nuclides can be quantitatively related to suitable gamma-emitters of similar origin. The method is tested by statistical analysis on the waste data recorded from two Swedish nuclear power plants. As this method is not applicable for Carbon-14, this nuclide was measured directly in spent ion exchange resins from three Finnish and Swedish power plants.

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1 INTRODUCTION

Nuclide content is one of the most significant factors affecting the optimization of procedures for reactor waste management. Insufficient knowledge of the activities of the most critical nuclides in reactor wastes can result in substantial economic losses; ultraconservative estimates can lead to oversophisticated waste management systems, whereas underestimation may necessitate later modifications of the waste management system.

At present concentrations of various nuclides in reactor wastes are insufficiently known. This is due to the lack of systematic measurement data of nuclide contents as well as to the lack of knowledge of generation of active nuclides and their subsequent release and transportation in reactor circuits.

During the last few years, attempts have been made to develop routines for activity measurement in wastes at Swedish and Finnish NPPs (Nuclear Power Plants). Monitoring equipment has been constructed, which is designed to meet the requirements laid down by the radiation protection authorities; to meet these requirements the activities of all significant nuclides must be determined in reactor wastes to be stored for later disposal.

The monitoring systems for reactor wastes used at Swedish and Finnish NPPs are based on gammaspectrometric measurements, thus they omit a number of pure beta emitters or weak gamma emitters. In order to obtain a complete understanding of nuclide content in wastes, the activity of such "problematic" nuclides needs to be determined - either directly or indirectly. In the present report the main emphasis is placed on indirect methods, where the activities of the problematic nuclides are determined by relating them to a few prevailing, gamma-emitting nuclides, which have analogous behaviour in reactor circuits with the nuclides to be determined.

In the present study, only those nuclides are considered, that are significant in reactor waste management. They are charaterized by relatively long halflives (about or more than one year), and at least one of the following properties: prevalent occurence in reactor wastes, high radiological toxicity, high tendency to escape from wastes and migrate in air, soil or rock medium. Chapter 2 provides a list of the significant nuclides including their relevant properties.

Chapter 3 summarizes a study of published literature on nuclide contents in reactor wastes. As the data are sparse and often incompletely reported, only a rough understanding of the prevalence of various nuclides is obtained. In this chapter the connection between activities in the primary coolant and wastes is also reported.

At Swedish NPPs, gamma-activities in practically all stored waste packages have been monitored by means of specially designed equipment. The comprehensive data files from Oskarshamn and Barsebäck NPPs were available for the study offering a firm base for systematic statistical analyses. Although only gamma-emitting nuclides are included in the analyses, the information obtained could probably be more widely applied to identify the factors contributing to variation ranges, correlations between nuclides etc. The analyses made are described in detail in ref /l/ and summarized in chapter 4.

The main objective of the study, development and evaluation of methods for determining the activities of nuclides which cannot be detected with present measurement techniques is dealt with in chapter 5.

Indirect methods can be used in order to determine the most problematic nuclides; this is feasible because of the distinct relations between nuclides of similar origin, as proved by the statistical analysis. The usefulness of existing models for generation, release and transportation of nuclides in reactor circuits is evaluated. For Carbon-14 a direct method of determination was selected, as its origin is quite different from the other nuclides considered. Experimental efforts to measure

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Carbon-14 in spent ion exchange resins from three NPPs are reported in detail in ref /2/.

This study did not include the extensive series of measurements which would be necessary for verifying the suitability of the models and determining the parameter values. If such work is to be performed, the present study may be a useful basis for identifying the shortfalls of the present knowledge. 2 NUCLIDES OF SIGNIFICANCE IN REACTOR WASTE MANAGEMENT

2.1 CORROSION PRODUCTS

Only those corrosion products, which possess quite high generation rate and relatively long halflife (about or more than one year), are of significance in reactor waste management. Those nuclides and their principal characteristics are summarized in Table 2-1.

Among the corrosion products listed only Iron-55, Nickel-59 and -63 are not gamma-emitters and thus cannot be detected from the outside of waste packages. It is therefore necessary to estimate their activity indirectly, for instance by relating their activities to the activities of certain gamma-emitting nuclides. It is evident that there are good grounds for such connections since Iron-55 has a gamma-emitting isotope Iron-59 with the same parent element whereas Nickel-59 and -63 have a common parent element with Cobalt-58, a gamma-emitter.

2.2 FISSION PRODUCTS

Table 2-2 summarizes the relatively long-lived $(T_{1/2} \gtrsim \text{one year})$ fission products, which are considered to have significance in management of reactor wastes. Some of their principal characteristics are summarized as well.

It is obvious that Ruthenium-106, Cesium-134 and Cesium-137 can be easily detected gammaspectrometrically, the detection of Cesium-144 is more difficult whereas Strontium-90, Technetium-99 and Iodine-129 cannot be detected at all and therefore their activities have to be determined by indirect methods. Because the nuclides given in Table 2-2 may be chemically quite different from one another, the existence of applicable relations between various nuclides is not as evident as in the case of corrosion products. Even in other respects, fission products form a more heterogeneous category than corrosion products; there are substantial differences in volatility, radiotoxicity, tendency to

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Table 2-1 The most significant corrosion products in reactor waste and their principal characteristics $(T_{1/2} \gtrsim 1 a)$

Nuclide	Half life	Parent Nuclide	Radiation mode	Chemical characteristics
Mn-54	0.83 a	Fe-54	strong gamma emitter	<pre>metallic, (amphoteric) dissolved (cation+ anion) and crud form</pre>
Fe-55	2.6 a	Fe-54	x-rays	metallic, crud form
Co-60	5.3 a	Co-59 Ni-60	strong gamma emitter	metallic, dissolved (cation) and crud form
Ni-59	8.10 ⁴ a	Ni-58	x-rays	metallic, dissolved (cation+anion)form
Ni-63	92 a	Ni-62	weak beta emitter	metallic, dissolved (cation+anion) form
3n-65	0.67 a	Zn-64	strong gamma emitter	metallic (amphoteric dissolved (cation+
Ag-110m	C.69 a	Ag-109	strong gamma emitter	anion) form metallic, dissolved (cation+anion) form

Table 2-2 The most significant fission products in reactor wastes and their principal characteristics $(T_{1/2} \gtrsim 1 a)$

Nuclide	Half life	Radiation mode	Chemical characteristics
Sr/Y-90	29 a	pure beta emitter	metallic, dissolved (cation) or crud form
Тс-99	2.1.10 ⁵ a	weak heta emitter	metallic (amphoteric) dis- solved (cation+anion) form
Ru/Rh-106	l.0 a	strong gamma emitter	metallic, crud or particulate form
I-129	l.7·10 ⁷ a	weak beta emítter	non-metallic (gaseous) dissolved (anion) form
Cs-134	2.1 a	strong gamma emitter	metallic, dissolved (cation) form
Cs-137	30 a	strong gamma emitter	metallic, dissolved (cation) form
Ce/Pr-144	0.78 a	weak gamma emitter	metallic, crud or particulate form

leach out of waste and in migration properties in soil and rock. Therefore it is not possible to give preferences to certain fission products according to their harm in waste management, but their relative significance is to a great extent dependent on the sequence of the events in question.

2.3 ACTINIDES

The content of actinides in the primary circuit is generally so low that they cannot be detected in primary water by routine methods. Radiologically all actinides are quite similar, all are strong alpha emitters except for Plutonium-241 which is a beta emitter. Their gamma and x-ray emissions are very weak. Because of their similarity and low concentrations it is not practicable to separate them so for this purpose they are treated as a group and their concentration is measured as Plutonium-239-equivalent.

As the amount of Uranium contamination on fuel surfaces is now insignificant, the main source of actinides in primary circuit is through major fuel damages. The release and transportation mechanisms of actinides are even less known, thus it is logical to estimate the activity of actinides in wastes by relating them to a suitable gamma-emitting fission product. As the actinides contribute only slightly to the risks associated with reactor waste management, quite rough, conservative estimates for their activities are adequate.

2.4 OTHER NUCLIDES

In addition to the nuclides already mentioned, at least Carbon-14 seems to deserve consideration in reactor waste management. Practically all Carbon-14 accumulated in reactor wastes originates from the Oxygen of the coolant or its Nitrogen-containing additives. Carbon-14 is relatively longlived, its halflife is 5730 years, and it emits only soft beta radiation.

Most of the Carbon-14 in coolant escapes through the gas purification system of the reactor. Carbon-14 contributes a substan-

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tial fraction of the long-term collective dose from LWRs: the integral over 500 years of the collective dose from the operation of LWRs has been assessed at about 0.025 manSv (2.5 manrem) per MW year, while the limit recommended by the Nordic radiation protection institutes for the total collective 500 year dose commitment is 0.01 manSv (1 manrem) per MW year /3/. Even though the amounts of Carbon-14 in reactor wastes are quite minor compared to atmospheric releases, it migrates rather rapidly in soil and rock and its dispersion there is poorer than in atmosphere, thus the risks from disposed reactor wastes may appear to be locally significant. 3 PUBLISHED DATA ON NUCLIDE CONCENTRATION IN REACTOR WASTES

3.1 WASTE ACTIVITY DATA

A review of available literature shows that the amount of waste activity data based on systematic measurements is very small. Nearly all the data published have rather conservative designbasis values, often with ill-defined experimental grounds. One cannot help noticing the striking discrepancies between data from various sources especially those concerning nuclides difficult to detect. The discrepancies can be partly explained by constructional and operational differences between various plants, but mostly they must be an indication of insufficient knowledge of activities in reactor wastes.

In ref /4/a study of the literature on activities of different nuclides in reactor waste was made. Table 3-1 summarizes the reported activities from six sources. For ease of comparison all the data have been normalized to correspond to the annual generation of reactor waste activity from an 1 GW(e) reactor (excluding core components). The age of waste was not well defined in every case, which makes the data for shortlived nuclides inaccurate in some cases.

Table 3-1 indicates that the activity data especially for Carbon-14, Nickel-59, Nickel-63, Strontium-90, Technetium-99, Iodine-129 and Plutonium-239 are either sparse of dispersed. This is evidently due to the fact that they are not gamma-emitters and therefore difficult to measure by routine methods. The dominating corrosion product nuclide seems to be Cobolt-60 in most cases, whereas Cesium-137 is generally the most significant fission product.

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Table 3-1 Reported annual activities in reactor wastes (core components excluded). All the data have been scaled to correspond to the nominal LWR output of 1 GW(e). Unit $TBq/GW(e) \cdot a$.

Nuclide	Half-life (a)	DOE/ET-0028 /5/	NUREG/CR-0680 /6/	NUREG-0782 BWR	/7/ PWR	Leddicote /8/	Nedo-10951 /9/	ALMA /10/
C-14	5730		0.0013	0.012	0.0020	0.058		$2.8 \cdot 10^{-4}$
Mn-54 Fe-55 Ni-59 Co-60 Ni-63 Zn-65	0.83 2.6 8.10 ⁴ 5.3 92 0.67	0.74 16 9.6	1.4 0.016 4.1.10 ⁻⁵ 7.6 0.088	17 0.017 28	1.0 0.0012 1.9	23	0.68 15 0.031	0.0057 30 0.50
Sr/Y-90 Tc-99 Ru/Rh-106 I-129 Cs-134 Cs-137 Ce/Pr-144	$29 \\ 2.1 \cdot 10^{5} \\ 1.0 \\ 7.10^{7} \\ 2.1 \\ 30 \\ 0.78$	12 0.074 74 70 0.18	$0.0032 - 5 \\ 1.0.10^{-6} \\ 2.1.10^{-6} \\ 2.6 \\ 4.8$	$0.044 \\ 7.5 \cdot 10^{-4} \\ 2.0 \cdot 10^{-3} \\ 20$	$0.0041 \\ 1.7 \cdot 10^{-5} \\ 5.0 \cdot 10^{-5} \\ 0.46$	0.2 0.063 0.0019 31	$\begin{array}{c} 4.2\\ 2.3\\ 3.7\cdot10^{-6}\\ 2.5\\ 4.4\\ 0.38\end{array}$	1 15
Pu-239	2.4.10 ⁴	0.10	$1.4 \cdot 10^{-5}$	$3.2 \cdot 10^{-3}$	$9.4 \cdot 10^{-4}$		0.50	3.7.10 ⁻⁵

Note: 1 TBq = 27 Ci

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3.2 RELATIONSHIP BETWEEN NUCLIDE CONCENTRATION IN WASTES AND IN PRIMARY CIRCUIT

Practically all the activities in reactor wastes have their origin in primary circuit, either directly or indirectly, therefore the nuclide composition in various reactor wastes should resemble the composition of nuclides generated in primary circuit. The strength of relationship is primarily dependent on the transportation mechanisms of nuclides from the source of activity to the waste material. An example of a direct transport route and accordingly strong relationship between source and waste might be reactor waste cleanup resins, whereas - as an opposite example - the transportation mechanisms from activity source to air filters are very selective and consequently the relationship between nuclide compositions is weak.

A demonstration of the distributions of nuclides in various water streams of the reactor is given in table 3-2. The activity data were measured at the St Lucie Plant, Unit 1 (2570 MW(t) PWR) /ll/. The consistency between various spectra of nuclides seems to be very good for corrosion products, whereas the correlations between fission products are much inferior; the ratio of Cesium-137 and Strontium-90 activity in various water streams vary by as much as two orders of magnitude. This discrepancy can probably be explained by the substantial differences in the physico-chemical properties of said nuclides.

Nuclide	Primary coolant (MBq/m ³)	Fuel pool (MBq/m ³)	Waste water system (kEq/m ³)
Mn-54	1.4	0.14	0.16
Co-60	27	2.6	3.7
Sr-90	1.3	0.013	0.20
Ru/Rh-106	1.2	0.12	0.16
Cs-137	1 600	1 600	250
Ce/Pr-144	20	21	2.3

Table 3-2 Measured activities in various water streams of the St Lucie Plant, Unit 1 (2570 MW(t) PWR) /11/

An illustration of the distribution of corrosion product nuclides in various parts of the primary circuit of an Asea-Atom type BWR is given in table 3-3. The data are based on calculations made by means of the computer code CRUD with an extensive empirical data basis. Again the consistency of various nuclide spectra seems to be reasonably good.

Table 3-3 Calculated activities in primary water, on surfaces of primary circuit and in reactor water cleanup resins after operating period of 20 a. Reference reactor Oskarshamn 2 (580 MW(e) BWR). /12/

Nuclide	Primary water (kBq/kg)	Surfaces of primary circuit in contact with water (MBq/m ²)	Ion exchange resins (TBq)
Mn-54	2.4	140	1.3
Fe-55	4.6	4 600	7.8
Co-60	3.5	850	11
Ni-59	0.0015	2.3	0.011
Ni-63	0.28	410	1.9
Zn-65	4.8	120	2.2

The third example originates from the Finnish Loviisa 1 reactor, which is a PWR of Soviet VVER-440 type. Table 3-4 summarizes the activities of three significant activation products in primary coolant, in evaporator bottoms reservoir and in effluents to the recipient /13/. Three sets of results according to the sampling date have been reported: the first around the turn of the year 1977/1978, the second 1978/1979 and the third 1979/1980 respectively. With few exceptions the correlations between various nuclides seem to be satisfactory, especially if it is taken into consideration that the origins of the samples cannot be compared.

Table 3-4 Measured activities of primary coolant, evaporator bottoms and controlled effluents to the recipient /13/. Reference reactor Loviisa 1 (440 MW(e) PWR of Soviet VVER-440 type). The samples were taken around the turn of the year a) 1977/1978 b) 1978/1979 and c) 1979/1980.

Nuclie	le	Primary coolant (kBq/m ³)	Evaporator bottoms, cumulative activity (MBq)	Controlled effluents (MBq/a)
	a)	190	1.5	_
Mn-54	b)	190	520	590
	c)	670	2 300	1 000
	a)	160	4.4	-
00-60	b)	8.5	590	410
	c)	14	4 000	1 300
	a)	110	48	-
Ag-110m	b)	48	1 600	280
	с)	44	20 000	5 600

In general the data published in open literature support the objectives of the study: the determination of the activities of the problematic nuclides by relating them to measurable ones. However the data available provide no sufficient basis for the determination of coefficients for such relations.

4 ACTIVITY DISTRIBUTION IN REACTOR WASTES AND CONTRIBUTORY FACTORS

4.1 STATISTICAL ANALYSIS OF WASTE ACTIVITY DATA

The activity distribution of certain nuclides in wastes is affected by several factors of both random and systematic character. Operating conditions, for instance, are mostly of random character even though they may have systematic features also. An example of a strong systematic influence on waste activities is the initial buildup of longlived nuclides on the surfaces of the primary circuit before reaching the productiondecay equilibrium.

If relatively long periods (several years) are considered, major random variations are expected to become averaged and thus statistical methods might offer a useful tool for visualizing the distribution of activities and correlations between various nuclides. As the distribution is spread over several orders of magnitude and several random factors affect it, the log-normal distributions are expected to be the most convenient simple models for describing the distributions. Deviations from lognormal distributions indicate the existence of strong systematic effects, dependencies between affecting factors or too infrequently occurring strong influences.

In /l/ a statistical analysis made on waste activity data obtained from Oskarshamn and Barsebäck power plants in Sweden, is presented in detail. As the analysis was restricted to a group of gamma-emitting nuclides - due to absence of data - it doesn't directly promote the objectives of the study. However, the analysis does provide background information on activity distributions and correlations between activities of various nuclides. Probably these distributions and correlations are not restricted only to the nuclides in question, but are more generally applicable for nuclides of analogous.

In the analysis, the specific activity in Bq/kg waste was first

calculated for each nuclide and for each unit of waste. The unit of wastes from Oskarshamn was equal to one package of waste (several types depending on waste) while the unit of waste from Barsebäck was one batch to be solidified (yielding several drums of waste). The units of waste were arranged in sequence according to their specific activities, and their cumulative distributions were plotted on probability paper (log-normal coordinates). The waste units were plotted according to the plant, the waste type and the nuclide. On most of the diagrams separate curves corresponding to different years of production were plotted in addition to the total curves. The log-normal fitting function (straight line) of the total curve was also plotted. Some examples of the distributions obtained are given in figures 4-1, 4-2, 4-3 and 4-4.

In order to test the correlations between various nuclides, a method was selected which was considered to be the most illustrative for the purposes of the study. The ratios of the specific activities of various nuclides were calculated and their respective statistical parameters were determined as for individual nuclides. The deviations of the distributions for ratios of nuclides and for separate nuclides were compared with each other. If there were no correlations the deviations of the ratios should be greater than the deviations of separate nuclides; the smaller the former are compared with the latter, the stronger is the correlation. Tables 4-1, 4-2, 4-3, 4-4 and 4-5 summarize the results of analyses made for various waste types. The figures obtained from both the actual distributions (measured) and the log-normal fitting curve (model) are presented. The variation ranges are given as the ratio between the 90 % quantile and the 10 % quantile.

Some attempts to test the suitability of the log-normal distributions were also made; these are described in greater detail in ref /1/.

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Fig. 4-1 Distribution of Cs-137 activity in concrete moulds containing granular ion exchange resins from Oskars-hamn NPP. Average dry waste content 40 kg/mould.



Fig. 4-2 Distribution of Co-60 activity in concrete moulds containing powder ion exchange resins from Oskarshamn NPP. Average dry waste content 200 kg/mould.



Fig. 4-3 Distribution of Mn-54 activity in batches of granular ion exchange resins from Barsebäck NPP. Average dry waste content 1200 kg/batch.



Fig. 4-4 Distribution of Zn-65 activity in batches of powder ion exchange resins from Barsebäck NPP. Average dry waste content 1500 kg/batch.

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Table 4-1 The statistical parameters characterizing the distributions of activities of separate nuclides as well as relative activities of nuclides. Activity unit MBq/kg dry waste.

Cemented gran	ular resins	from	Oskarshamn,	average	dry	waste	content	40	kg/	/packad	дe
---------------	-------------	------	-------------	---------	-----	-------	---------	----	-----	---------	----

Nuclide or relation between	l0 % quantile		Median		Average measured	90 % quantile		90 % quantile/ 10 % quantile		Number of	ar - Land and a company of the second se
nuclides	measured	model	measured	model	& model	measured	model	measured	model	tions	
Nuclide									······································		}
Mn-54	9.7	9.1	53	57	150	470	300	48	39	69	
C0-60	240	290	2 600	1 900	3 800	8 400	12 000	35	40	266	1 1
Zn-65	420	290	1 800	1 500	2 700	5 600	7 400	13	25	253	9
Cs-134	130	110	1 100	1 200	6 600	26 000	14 000	200	130	234	
Cs-137	230	190	2 100	2 300	14 000	47 000	27 000	200	140	259	
Relation											
Mn-54/Co-60		0.0022		0.062	0.086		0.17		7.7	69	
Zn-65/Co-60		0.18		0.66	1.0		2.4		13	253	
Cs-134/Cs-137		0.20		0.48	1.1		1.2		6.0	233	ł
Co-60/Cs-137		0.20		0.92	2.3		4.2		21	259	- - -
											l J

Note: $I MBq = 2.7 \cdot 10^{-5} Ci$

Table 4-2 The statistical parameters characterizing the distributions of activities of separate nuclides as well as relative activities of nuclides. Activity unit MBq/kg dry waste.

Cemented powder resins from Oskarshamn, average dry waste content 200 kg/package

Nuclide or relation	10 % quantile		Median		Average measured	90 % quantile		90 % quantile/ 10 % quantile		Number of observa-	
nuclides	measured	mode1	measured	model	model	measured	model	measured	model	tions	
Nuclide											-1
Mn-54	0.036	0.031	0.24	0.20	0.56	1.1	1.3	31	42	85	
C0-60	0.23	0.23	2.4	2.1	8.1	15	19	65	80	246	
Zn-65	0.072	0.068	0.74	0.81	5.0	8.9	9.8	120	140	226	
Cs-134	0.13	0.10	1.5	2.0	17	52	39	400	380	233	
Cs-137	0.12	0.11	2.8	2.8	34	110	77	920	730	313	
Relation											i i
Mn-54/Co-60		0.017		0.044	0.074		0.12		6.9	85	
Zn-65/Co-60		0.055		0.18	0.38		0.57		10	226	ļ
Cs-134/Cs-137		0.15		0.28	0.35		0.52		3.6	227	
Co-60/Cs-137		0.17		0.83	2.7		4.1		24	310	
											i

Note: $I MBq = 2.7 \cdot 10^{-5}$ Ci

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Table 4-3 The statistical parameters characterizing the distributions of activities of separate nuclides as well as relative activities of nuclides. Activity unit MBq/kg dehydrated waste.

Dehydrated powder resins from Oskarshamn, average waste content 6 m³/package.

Nuclide or relation between nuclides	10 % quantile		Median		Average measured	90 % quantile		90 % quantile/ 10 % quantile		Number of	
nuclides	measured	model	measured	model	model r	measured	model	measured	model	tions	
Nuclide											1
Mn-54	0.19	0.19	0.50	0.51	0.69	-	1.4	-	7.6	37	
Co-60	2.4	2.6	9.7	8.8	13	24	30	10	12	37	2
Zn-65	0.45	0.45	1.4	1.4	2.1	5.1	4.5	11	10	37	, <u> </u>
Co-58	0.24	0.32	1.7	1.7	3.3	8.3	8.6	35	27	37	
Ag-llOm	-	0.12	0.31	0.33	0.45	1.1	0.90	-	7.7	37	1
Relation											
Mn-54/Co-60		0.024		0.066	0.11		0.18		4.3	37	:
Zn-65/Co-60		0.073		0.18	0.22		0.43		5.9	37	
Co-58/Co-60		0.078		0.21	0.35		0.58		7.4	37	1
Ag-110m/Co-60		0.014		0.042	0.097		0.13		9.6	37	
											ł

Note: $1 \text{ MBq} = 2.7 \cdot 10^{-5} \text{ Ci}$

Table 4-4 The statistical parameters characterizing the distributions of activities of separate nuclides as well as relative activities of nuclides. Activity unit MBq/kg dry waste.

Bituminized granular resins from Barsebäck, average dry waste content about 1200 kg/batch

Nuclide or relation between	l0 % quantile		Medi	an	Average measured	90 % quantile		90 % quantile/ 10 % quantile		Number of	-
nuclides	measured	model	measured	model	& model	measured	model	measured	model	tions	
Nuclide								•			
Mn-54	0.29	0.32	16	6.5	32	94	130	320	410	34	
Co-60	2.7	2.7	160	73	390	960	2 000	360	750	3.6	
Zn-65	0.62	0.94	66	19	93	240	390	390	410	35	N.
Co-58	0.49	0.54	13	17	180	360	560	730	1 000	37	
Ag-llOm	-	0.21	16	5.5	32	78	130	-	610	34	1
Relation											1
Mn-54/Co-60		0.055		0.078	0.084		0.11		2.1	34	6 4
Zn-65/Co-60		0.15		0.22	0.24		0.33		2.2	35	
Co-58/Co-60		0.10		0.27	0.46		0.72		7.2	36	ł
Ag-110m/Co-60		0.022		0.053	0.078		0.13		5.9	34	

Note: $1 \text{ MBq} = 2.7 \cdot 10^{-5}$ Ci

Table 4-5 The statistical parameters characterizing the distributions of activities of separate nuclides as well as relative activities of nuclides. Activity unit MBq/kg dry waste.

Bituminized powder resins from Barsebäck, average dry waste content about 1500 kg/batch

Nuclide or relation between nuclides	l0 % quantile		Median		Average measured	90 % quantile		90 % quantile/ 10 % quantile		Number of observa-	
	measured	model	measured	model	& model	measured	model	measured	model	tions	
Nuclide											
Mn-54	-	0.056	0.21	0.23	0.37	0.86	0.91	-	16	42	1
Co-60	1.6	1.5	4.1	3.7	4.5	8.1	9.0	5.1	6.0	59	1
Zn-65	0.98	1.0	3.6	3.3	4.6	11	11	11	11	59	ω
Cs-134	0.16	0.17	1.0	1.1	2.9	5.8	7.1	36	41	50	1
Cs-137	0.22	0.21	1.4	1.8	6.2	14	15	64	72	55	
Relation											
Mn-54/Co-60		0.023		0.057	0.081		0.14		6.1	42	
Zn-65/Co-60		0.40		0.89	1.2		2.0		4.8	59	ĺ
Cs-134/Cs-137		0.29		0.52	0.62		0.94		3.2	48	
Co-60/Cs-137		0.46		2.2	6.0		11		23	59	f :

Note: $1 \text{ MBq} = 2.7 \cdot 10^{-5} \text{ Ci}$

4.2 CONCLUSIONS FROM THE STATISTICAL ANALYSIS AND DISCUSSION OF THE CONTRIBUTORY FACTORS

Some preliminary conclusions from the results of the statistical analysis are given below together with some ideas of the factors that contribute to the deviations.

- 1. The specific activities of the corrosion products (Manganese-54, Cobalt-60 and Zinc-65) in powder ion exchange resins from the two plants are fairly close to each other, while the corrosion product activity of granular ion exchange resins from Oskarshamn is about ten times greater than that from Barsebäck. This is probably due to differences in the cleanup systems at the plants. At both plants the nuclide composition is similar in granular and powder ion exchange resins, whereas a comparison shows that at Oskarshamn Zinc-65 is by far more dominating than at Barsebäck. This is probably an indication of differences in construction materials of the primary circuits of the plants.
- 2. Cesium-137 and Cesium-134 in the wastes from Oskarshamn were the only fission products encountered. The ratio of the average specific activities of Cesium in granular and powder resins is aroung 400 which is about the same as for corrosion products. The dispersion of activity data for Cesiums is significantly wider than for corrosion products: the scattering of data measured by the 90 % quantile/the 10 % quantile is between 100 and 1000. This is due to burst releases involved with some fuel failures: for long periods there may be practically no fission products in the primary circuit then as a consequence of major fuel failures they will rapidly become a dominating source of relatively longlived activity.
- 3. The correlations between various corrosion products can be tested through the statistical parameters of the ratios of their activities. They can be summarized as follows:

Statistical		Relative activities			
parameters	NPP	Mn-54/Co-60	Zn-65/Co-6 0		
Average	Oskarshamn	0.0740.086	0.381.2		
	Barsebäck	0.0840.11	0.220.24		
Variation	Oskarshamn	6.17.7	4.813		
range *	Barsebäck	2.14.3	2.25.9		

Table 4-6 Summary of statistical parameters for relations between corrosion products.

*The variation ranges are expressed as the 90 % quantile/10 % quantile.

It appears that Manganese-54 is more closely related to Cobalt-60 and that the coefficient of this relation is quite independent of the plant. The relation of Zinc-65 to Cobalt-60 is looser and dependent on the plant (as concluded before). The variation ranges at Oskarshamn are 5...13 and 2...6 at Barsebäck. The differences between the plants can be explained by the differences in size of the waste units. The variation ranges for relative activities are quite moderate compared with those of absolute activities (up to several hundred at both Oskarshamn and Barsebäck). This supports the theory of quantitative relations between nuclides of similar origin and relatively similar chemical characteristics.

4. The only fission products included in the analysis were Cesium-137 and Cesium-134, which have unlike origin, but quite idential release and transportation properties, consequently their correlation was found to be quite close: the 90 %/10 % variation range between 3...6. It cannot be anticipated that the correlations between fission products are generally as close.

5. There was no strong correlation between the specific activity

of waste and the operating age of the reactor. This indicates that the variations are mainly due to random factors, such as fuel failures, operating details etc.

6. Statistical tests indicated that the distributions differ significantly from purely log-normal distribution, as expected. However, examination of the curves shows that most of the lognormal lines agree fairly closely with the measured curves, which was also confirmed by some quality tests. The distributions of the activities of granular resins from Barsebäck turned out to be the most curved. A survey of the annual distributions shows that the batches are divided into quite separated groups, which causes the skewness. Probably this skewness tends to decrease when more batches of waste are included in the analysis.

Although the factors affecting distributions of waste activities do not in every respect fulfill the requirements of lognormality, it is obvious that log-normal distributions provide a useful tool where activity distributions in various items of waste are of importance. The distribution can then be defined by means of only two parameters: the median (the logarithmic mean) and the logarithmic standard deviation.

5 RELATIONS BETWEEN GENERATION, TRANSPORTATION AND ACCUMULATION CHARACTERISTICS OF VARIOUS NUCLIDES

5.1 MODELS FOR ACTIVATION PRODUCTS

The principal mechanism for the build-up of corrosion product activity in wastes is the following chain: corrosion of surfaces in the primary circuit -> release from surfaces and transportation into the core -> deposition on surfaces in the core -> activation of core materials or corrosion products in coolant or in deposits on core surfaces -> release of activated nuclides in the core and transportation into reactor circuits -> removal of active corrosion products by filtration, ion exchange or by leakages. There are several analytical models for both PWRs and BWRs which describe quantitatively the processes mentioned above. They are mainly intended for reactor design, radiation dose reduction or decontamination simulation, but it appears that they are in principle also applicable to estimates of the build-up of activity in wastes originating from ion exchange, filtration or by leakages.

Of course, calculation of activity accumulation in wastes by such models is not a straightforward task. The models utilize a large set of parameters which, besides being reactor-specific also tend to be sensitive to the operating conditions of the reactor. In addition, most of the models fail in the proper simulation of corrosion products bursts after power transients of the reactor, which can essentially alter the activity distributions in the primary circuit. Thus, the models for corrosion product transportation provide no easy way for the direct calculation of activities in wastes originating from the primary circuit, but their value becomes apparent when searching for methods to indirectly determine the activities of the problematic nuclides in wastes.

In the study, the value of three corrosion transportation models was submitted to detailed examination. Two of them are of Swedish origin and are applicable to BWRs /12, 15/, the third was of Hungarian origin and was particularly adapted to the Finnish

Loviisa-l reactor during the scholarship period of the author of the code in Finland /16/.

The CRUD code of Asea-Atom possesses a firm data basis through the extensive radiochemical data generating program of Asea-Atom at Swedish and Finnish BWRs. As an example of the applicability of the model, the results of a simulation of a 20-year history of six important nuclides in waste management have been presented in ref /12/. Oskarshamn-2 was used as a reference reactor for the calculations. The nuclide concentrations that were accumulated in various parts of the primary circuits at the end of the 20-year period are given in table 3-3. Figure 5-1 is an example of a graphic presentation of the cumulative activities of two nuclides in reactor water cleanup resins. The differences in accumulation rates due to the unequal halflives of the nuclides are clearly shown.

Another Swedish model for corrosion product behaviour in BWR elaborated at Studsvik, is a constant-coefficient block model, designed primarily for the simulation of crud buildup on the surfaces of the primary circuit. However, it is also applicable to calculating the accumulation of corrosion products in ion exchange resins. In ref /15/ a demonstration calculation for Iron-59 is presented. A disadvantage of the model, compared with the CRUDmodel, is that at present there is not sufficient experience to establish its validity for power reactors.

The third model described in ref /16/ is the RADTRAN, an improved version of the RADSYS code originally developed in Hungary. The code has been specially applied to the Loviisa-1 PWR of the Soviet VVER-440 type, but in principle it is applicable to any PWR and even BWR because of its flexible graph-theory techniques in describing the units of the primary circuit and connections between them. The code is able to simulate a wide range of operational modes of a typical PWR, and it is suitable for the calculation of fluid, surface and resin activities in the primary circuit. In its present form the code suffers from lack of measurement data for input parameters and for testing the model.

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Fig. 5-1 Cumulative activities of Cobalt-60 and Nickel-63 in reactor water cleanup resins during operating period of 20 a. Reference reactor Oskarshamn-2. Simulations were made by means of the CRUD-code of Asea-Atom /12/.

1

As indicated in chapter 2.1, Iron-55, Nickel-59 and -63 were the only corrosion products which could not be measured by direct routine methods from packages of wastes, and it is necessary to determine their activities by indirect methods. A survey of the list of nuclides shows that by origin they are closely related to certain gamma-emitters; Table 5-1 illustrates the situation:

Table 5-1	Properties	relevant	to the	generation	. rate of	certain
	activation	products	having	analogous	origin.	

Parent	Reaction	Daughter	Half-life	Isotopic	Cross section
			(a)	fraction (%)	(m ² /kg)
Fe-54	(n,p)	Mn-54	0.86	5.8	5.2.10 ^{-6*}
Fe-54	(n , ¥)	Fe-55	2.7	5.8	1.8.10 ⁻⁴
Fe-58	(n ,%)	Fe-59	0.12	0.31	4.1.10 ⁻⁶
Ni-58	(n,p)	Co-58	0.20	68	7.9.10 ^{-5*}
Co-59	(n ,\$)	Co-60	5.3	59	0.039
Ni-58	(n ,¥)	Ni-59	80 000	68	3.3.10 ⁻³
Ni-62	(n, \$)	Ni-63	92	3.7	6.1.10 ⁻⁴

*Cross section for fast neutron reaction.

Judging from their origins, Fe-59 and Co-58 might be the most suitable reference nuclides for the indirect determination of Iron-55, Nickel-59 and -63. However, they have one great disadvantage: because of their relatively short half-life they have often decayed to an insignificant level by the time the activity in wastes is measured. Thus Manganese-54 in relation to Iron-55, and Cobalt-60 in relation to Nickel-59 and -63 seem to be more representative reference nuclides. The relatively similar chemical characteristics of the said elements ensure that the relation varies within reasonable limits even though the reference elements are not idential with the nuclides to be determined. However, on the basis of present knowledge it is not possible to estimate the confidence limits for such relations. Models of corrosion product transportation in the primary circuit (examples of which are given above) provide a useful tool for the determination of the relations between various corrosion products.

5.2 MODELS FOR FISSION PRODUCTS

The main transportation route of fission products to wastes is the following chain: generation in fuel -> release from fuel to primary water -> transportation to various parts of reactor circuits -> removal by filtration, ion exchange or by leakages.

The first stage, generation in fuel, is comparatively well known: the burnup of fuel usually gives sufficient information for the estimation of its fission product and actinide inventory. Nowadays there are several advanced codes for the calculation of fission product inventories of which the ORIGEN is probably the best known.

The release of fission products from fuel can be divided into two sequential main phases: release from the fuel matrix and release through cladding. The main mechanisms leading to the release of fission products from the fuel matrix can be identified as the recoil release, the knockout release and the migration. Practically all the release through cladding originates from defective fuel elements, if gaseous fission products noble gases and iodines - are excluded.

Factors affecting the release from the fuel matrix depend essentially on the mechanism concerned. The recoil process is dependent solely on fission rate, while the knock-out process depends on both fission rate and fission product inventory. The migration process is nearly directly proportional to nuclide concentration in fuel: it has strong dependence on the temperature of the fuel and it is substantially affected by the condition of the fuel and the operation mode of the reactor.

Below linear power rating of 20 kW/m the recoil and knock-out processes dominate, whereas above 25 kW/m the release is almost entirely due to migration.

Cladding failures are generally caused by manufacturing defects, internal hydriding, accelerated corrosion, pellet-cladding interactions or mechanical damages. The trapping of fission products on the cladding has an important role in the clad escape mechanism. During power transients the trapped fission products may be completely released. The inclination of certain nuclides to be released after such transients is described by so called spiking factors. Due to delay effects the spiking factors are different for various isotopes having different half-lives. By the investigation of spiking factors the type and the location of cladding failures can be estimated.

Transportation of fission products in the primary circuit is not so complicated a problem as in the case of corrosion products. In the absence of a high concentration of inactive isotopes (contrary to corrosion products) most fission products are highly soluble. The principal removal mechanism of fission products in coolant is by cleanup systems, and only a minor fraction is expected to deposit on surfaces of the primary circuit. A prerequisite for solving the dependence between the fission product activities in coolant and in cleanup resins or filters is the determination of the efficiency (decontamination factor) or water purification systems. Generally this is not a straightforward task, as the cleanup efficiencies are dependent on species concerned, concentrations, pH, temperature and other operating conditions of the reactor. Proportionality between concentrations of various nuclides in coolant and in cleanup resins or filters is valid only provided that the decontamination factors for those nuclides are quite high.

A traditional approach to describing the release of fission products from fuel are the diffusion models, semi~empiric models which to some extent describe the physical processes involved.

They are best suited for low and medium temperatures and for noble gases, but fail especially in describing the releases from defective fuel elements during and after power transients. Thus they are not very useful as far as the present study is concerned.

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Recently, some models for fission product release have been developed, which achieve a better distinction between the various processes involved. Perhaps the most advanced of these is the PROFIP-code /14/. This code has been successfully applied in estimating the number and characteristics of fuel failures by means of isotopic ratios of released nuclides.

The RADTRAN code described in the previous chapter can also simulate fission product release and transportation in reactor circuits /l6/. In its present form its fission product release model is of the diffusion type and quite rough and thus not very practical for the purposes of this study. Due to its flexible structure, however, the model possesses a good development potential.

The problem area treated in the present study is in certain respects simplified compared with the application ranges of abovementioned models; such simplifications are:

- the nuclides concerned are relatively longlived, and accordingly their activities can be assumed to be constant during the release and transportation processes
- primary interest is laid on integral activity during relatively long operating periods
- only the relative proportions between nuclides are of interest
- the accuracies strived for are not very ambitious.

There seem to be two main alternatives for the construction of relations between fission products (and actinides) in wastes. They can either be based on measurements of fission product activities in primary water, or on estimates of fission product releases from fuel.

The first method has a few disadvantages. As discussed earlier, the decontamination factors are dependent on species, concentrations and other factors, therefore the relative fractions of the various nuclides released in coolant and in wastes are not the same or even constant. Measurements indicate that both the activities of separate fission products and their relative

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activities in primary coolant fluctuate rapidly. Measurements made at Oskarshamn /17/ showed that the activity of Cesium-137 in primary coolant can vary within two orders of magnitude and the ratio between Cesium-137 and Strontium-90 in primary coolant varied between about 10 and 10 000 depending on fuel performance and the operating mode of the reactor. Cesium seemed to have a very strong spiking tendency while Strontium had practically none. As the instantaneous activities in primary water can vary so widely, the determination of the relations based on only a few measurements of the "problematic" nuclides in primary coolant does not appear to be a dependable method.

The second method, estimation of fission products and actinides released from fuel, suffers from the disadvantage of the great dependence of release fractions on the process in question. However, it is possible to indentify the release mechanism involved by measuring in primary water the relative activities of certain short-lived Xenon and Iodine isotopes with different halflives. These methods, primarily intended for characterization and location of fuel failures, could also be suitable for determining the relation between fission product activities in wastes. The method suggested would be based on the determination of two or three relations depending on the release type dominating during the accumulation period of activities. Determination of such relations requires utilization of advanced models for fission product release and transportation as well as the completion of experimental programs at reactors during periods of fuel failure.

On the basis of present knowledge, table 5-2 gives a summary of the inventory in fuel and the leakage coefficient from fuel relative to Cesium-15: for certain non-gamma-emitting fission products and actinides as well as the average activity in wastes relative to Cesium-13: estimated by means of the previous figures.

The exceptionally wide variations of leakage coefficient from fuel for Strontium-90 and Plutonium-239 are due to the fact that they seem to have a weak tendency to "spiking leakage" from fuel, in contrast to the rest of nuclides in table 5-2.

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Table 5-2 Inventory in fuel, estimated range of leakage coefficient from fuel and estimated average activity in wastes relative to Cesium-137 for certain nongamma-emitting fission products and actinides.

Nuclide	Activity in fuel	Estimated range	Estimated average
	relative to Cs-137	of leakage coeffi-	activity in wastes
		cient relative	relative to Cs-137
		to Cs-137	
Sr-90	0.7	$10^{-3} \dots 0.2$	5.10 ⁻³ 0.1
Tc-99	$1.4 \cdot 10^{-4}$	0.11	$2.10^{-5} \dots 10^{-4}$
I-129	$3.4 \cdot 10^{-7}$	120	$3.10^{-7} \dots 5.10^{-6}$
C s- 135	2.7.10-6	1	3.10-6
Pu-239	3.0.10-3	$10^{-5} \dots 10^{-2}$	$10^{-7} \dots 10^{-5}$

5.3 MODELS FOR CARBON-14

In LWRs the main sources of Carbon-14 are the (n, α) - reaction of the Oxygen-17 of coolant and the (n, p) - reaction of the Nitrogen-14 dissolved in coolant; releases from fuel can be neglected. The reported amounts of Carbon-14 originating from coolant are around 200 GBq (5 Ci) per GW(e).a for PWRs and 300 GBq (8 Ci) per GW(e).a for BWRs /18/, which can be confirmed by simple theoretical calculations. There are some uncertainties associated with the Carbon-14 production of PWRs, as chemicals containing Nitrogen (NH₄OH, N₂H₂) can be significant sources of Carbon-14 and their amounts can vary within specified limits.

It is generally accepted that Carbon-14 produced in coolant almost totally escapes through the offgas-system of the reactor. A minor fraction of Carbon-14 is trapped by the water clean-up system and passes through the waste treatment system, from where part of it may be released. In the available literature there are only vague indications regarding the size of this fraction and its chemical characteristics.

In the view of the present attention directed towards Carbon-14,

an experimental program for Carbon-14 determination in wet wastes was carried out in the study; ref /2/ gives a detailed description of the program. The measurements were performed at two laboratories, at Otaniemi in Finland and at Studsvik in Sweden. Samples of ion exchange resins and primary water were obtained from three power plants: Loviisa (PWR) in Finland, Olkiluoto (BWR) in Finland and Oskarshamn (BWR) in Sweden. The objectives of the measurements were to find out the activity balance in equilibrium and the chemical form of Carbon-14 in the primary circuit.

The results indicated, as expected, that there are substantial differences in the transportation and chemical form of Carbon-14 depending on reactor type. At PWRs the conditions are highly reducing, thus a major portion of Carbon-14 appears as hydrocarbons or carbon monoxide, and a minor portion (30 % in the measurements) as carbon dioxide. At BWRs practically all Carbon-14 (88...98 %) was found as carbon dioxide. These results were obtained from samples of fresh ion exchange resins, while CO_2 was completely dominating in spent resins. Judging from this, hydrocarbons and CO are released from resins much more easily and consequently only the Carbon-14 as CO_2 needs to be considered for reactor waste management.

There were considerable differences in the amounts of Carbon-14 accumulated in resins depending on reactor type. These differences can be explained by the dissimilarity of the processes: the escape of Carbon-14 in gaseous form is increased by the steam phase flow of BWRs compared with the PWR process. Table 5-3 summarizes the amounts of Carbon-14 in reactor water clean-up resins for the three plants. The accumulation rate in resins for the PWR (Loviisa) was about 1.0 GBq (30 mCi) per GW(e) and for the BWRs (Olkiluoto and Oskarshamn) around 0.15...0.40 GBq (5...10 mCi) per GW(e) a. It is expected that these figures are good estimates for Carbon-14 content in all wet wastes, as the Carbon-14 accumulation in other wet wastes is supposed to be marginal. On the other hand air filters, especially charcoal filters, can contain considerable amounts of Carbon-14. As Carbon has quite a high tendency to escape from wastes, the final amount of Carbon-14 in disposed wastes can be much lower than in untreated wastes. For instance, during the drying phase of bituminization Carbon-14 is anticipated to be discharged almost totally. It is difficult to take such effects into account but fortunately their omission gives a conservative approach to the risk analysis of reactor waste management.

Table 5-3 Summary of the activity of Carbon-14 in reactor water clean-up resins from three Finnish and Swedish NPPs.

	Loviisa l	TVO I	Oskarshamn l
		(Olkiluoto)	
	PWR	BWR	BWR
Nominal output (MW(e)	440	660	450
Estimated C-14 produc-	60150	100200	100200
tion in primary			
coolant (GBq/a)			
C-14 activity in reactor	0.46	≤ 0.17	2 0.07
water clean-up resins			
(GBq/a)			
2-14 activity in resins/	C.30.8	0.050.1	0.050.1
C-14 produced in coolant			
(%)			

Note: 1 GBq = 27 mC1

5 CONCLUSIONS AND DISCUSSION

The experiences gained in the course of the study can be summarized in the following brief conclusions:

- . The monitoring technique currently in use at Swedish and Finnish NPPs makes it possible to detect all the significant gamma-emitting nuclides from packages of wastes. These measuring systems, however, omit several non-gamma-emitters of significance in reactor waste management, and it is necessary to determine their activities either by direct or indirect methods.
- 2. The data in available literature provide no adequate basis for estimation of the activities of those nuclides difficult to detect, since the discrepancies between data from various sources are strikingly large. In addition, activities in wastes seem to be strongly dependent on the type of reactor and its operating conditions.
- 3. Statistical analyses as well as published data indicate that there is good correlation between the activities of various nuclides in wastes and water streams of reactor circuits, provided that the nuclides in question have analogous origin and relatively similar chemical characteristies. In this respect corrosion products seem to be a more homogeneous group than fission products. From this it can be deduced that one possible way to determine the activities of the "problematic" nuclides in wastes is by relating them to the activities of certain gamma-emitting reference nuclides with analogous properties; the activities of the reference nuclides could be measured by means of the techniques referred to above.
- 4. There is a variety of computer codes for simulation of generation, release and transportation of corrosion products as well as fission products in reactor circuits. In Sweden and Finland there are three codes available, which are mainly intended for

the simulation of corrosion product behaviour in reactor circuits.

Determination of the relations between corrosion products seems to be relatively easy by means of the existing models and a few series of measurements made at the reactor in question. In many cases satisfactory estimates of the relations are obtained even if the dependency on reactor age and other operating conditions are neglected. In the case of fission products the problem is more difficult. Identification of the dominating release mechanism from fuel is probably needed and application of more complex relations necessary in order to attain sufficient accuracy. In their present form the existing models for fission product behaviour in reactor circuits are not sufficiently developed to provide the information necessary for determining the relations between nuclides. Thus, their further development based on extensive experimental verification is desirable.

5. In the study special emphasis was given to Carbon-14 as the knowledge of its behaviour in reactor circuits was recognized to be not sufficient. The measurements indicated that, depending on reactor type, 0.05...1 % of all Carbon-14 generated in coolant was absorbed by ion exchange resins, and that practically all this Carbon-14 was found as carbonates. A rough estimate of Carbon-14 content in reactor wastes can be made on the basis of the measurements.

As a conclusion it can be stated, that there are no easy solutions for determining the activities of those nuclides in reactor wastes which cannot be detected by the present monitoring techniques. Further development of the theoretical basis as well as experimental verifications of the behaviour of active nuclides in reactor circuits will be necessary as a complement to the routine activity determinations now performed.

It should be carefully considered, weighing the benefits against the efforts needed, whether there is sufficient justification for

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such additional work. One crucial criterion in decision making is the importance of the role which those "problematic" nuclides play in the safety analyses of reactor waste management.

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APPENDIX

Working documents belonging to the Nordic study on reactor waste

Appendix

Working documents belonging to the Nordic study on reactor waste

Documents are available from the following libraries:

Denmark:	Forsøgsanlæg Risø
	Postboks 49
	DK-4000 Roskilde
Finland:	Technical Research Centre of Finland
	Vuorimiehentie 5
	SF-02150 Espoo 15
Sweden :	Studsvik Energiteknik AB
	S-611 82 Nyköping
Norway	Institutt for energiteknikk
NOLWAY.	Dootholo 40
	POSTDOKS 40
	N-2007 Kjeller

Chronological list of NKA/AO documents:

NKA/AO-A:1 Pre-project report, November 1977.

- AO(78)1 System and risk analysis of reactor waste. Status per March 1. 1978. F. Marcus. 1978-03-07. (In Danish and Swedish).
- AO(78)8 Half-year report, 1st half-year 1978. 1978-08-08. (In Swedish).

AO(78)13 System and risk analysis of reactor waste. Proposal for the 1979 program. B. Berlin. 1978-12-08. (In Swedish).

AO(78)14 Product specifications. P.O. Nielsen. 1978-08-24. (In Norwegian).

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- AO(78)16 Water qualities. P.O. Nielsen. 1978-10-05. (In Norwegian and Swedish).
- AO(78)17 System description. Proposal for classification. P.O. Nielsen. 1978-12-18. (In Norwegian).
- AO 78)18 Some views on system analysis concerning barrier philosophy and planning of waste repository. K. Brodersen. Risø, December 1978. (In Danish).
- AO(78)19 Some general rules concerning the location of a final repository above ground in Denmark. K. Brodersen. Risø, December 1978. (In Danish).
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