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Radionuclides Difficult to Measure in Waste Packages Final Report

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# Radionuclides Difficult to Measure in Waste Packages Final Report

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# Final report on "Radionuclides Difficult to Measure in Waste Packages"

The enclosed report is the result of a project conducted jointly by the Swedish Nuclear Power Inspectorate (SKI), the Swedish Radiation Protection Institute (SSI) and the Swedish Nuclear Fuel and Waste Management Co (SKB). The report will not be published formally as a SKI Report, but it is distributed to a limited number of the receivers of such reports.

Yours sincerely,

yefors Stig-Wingefors

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# **Introduction**

This report has been prepared as part of the research project that Brenk Systemplanung is performing for the SSI (Swedish Radiation Protection Institute), the SKB (Swedish Nuclear Fuel and Waste Management Company) and the SKI (Swedish Nuclear Power Inspectorate). This work is a continuation of a first phase that was completed in 1992 [THI 92] in which nuclide specific data for the correlation between Ni 63 and Co 60 and measurement methods for nuclides difficult to measure were investigated.

In the present phase 2 correlation analyses for a variety of nuclides that are difficult to measure have been performed. The nuclides to be investigated have been chosen by the orderer. The choice was based on the results of the first phase of this project [THI 92] and on the special requirements for the Swedish waste streams as identified by the orderer.

According to the specifications of the orderer, the relevant nuclides are the following:

H 3, C 14, Cl 36, Ca 41, Ni 59, Ni 63, Sr 90, Mo 93, Tc 99, I 129, Cs 135, U 234, Np 237, Pu 238, Pu 239/Pu 240, Pu 241, Am 241.



# 1. Object and Scope

# **1.1.** Object of the Study

In this study nuclide specific correlation analyses between key nuclides that can be easily measured and nuclides that are difficult to measure are presented. Data are taken from studies and data compilations from various countries. The results of this study can serve to perform assays of the nuclide specific radionuclide contents in waste packages by  $\gamma$ -measurements of Co 60 and Cs 137 and calculation of the contents of the other nuclides via the correlation analyses, sometimes referred to as 'scaling factor method'. It can thus be avoided to have to take samples from the waste for separate analysis.

The attempt is made to also investigate the physical and chemical backgrounds behind the proposed correlations. For example, a formation pathway common to the two nuclides to be correlated can be regarded as an explanation, if a good correlation has been found. On the other hand, if the observed correlation is of poor quality, reasons may possibly lie in different behaviour of the two nuclides in the water system of the nuclear power plant. This implies not only chemical solubility, transfer constants etc. in the water system, which would not only affect the proportionality between the two nuclides, but a different behaviour in different parts of the water system must be assumed (e.g. different filter efficiencies etc.).

# 1.2. Situation in Sweden

In Sweden, only light-water reactors (LWR) are in operation at the moment. There are 3 pressurized water reactors (PWR) and 9 boiling water reactors (BWR) according to table 1. This justifies a limitation of the scope of this study to wastes from LWRs only. In addition, as the fuel cycle in Sweden can be characterized as 'once-through', only the nuclide vector for normally enriched U fuel need to be regarded for the transuranic (TRU) radionuclides and fission products.



# Table 1 Nuclear power plants in Sweden

Unit	Туре	Power rating	Start of construc-	Synchronisation
		MWe	Search tion	to the grid
Ringhals-2	PWR	905	1970	1974
Ringhals-3	PWR	960	1973	1982
Ringhals-4	PWR	960	1973	1982
Sum	3 PWR	2825		
Barsebäck-1	BWR	615	1971	1975
Barsebäck-2	BWR	615	1973	1977
Forsmark-1	BWR	1006	1971	1980
Forsmark-2	BWR	1006	1973	1981
Forsmark-3	BWR	1197	1977	1985
Oskarshamn-1	BWR	462	1966	1971
Oskarshamn-2	BWR	630	1969	1974
Oskarshamn-3	BWR	1205	1980	1985
Ringhals-1	BWR	825	1969	1974
Sum	9 BWR	.7561	· · · · · · · · · · · · · · · · · · ·	
Total	12 units	10386		

# **1.3.** Results from Previous Investigations

The investigation of the possibility to determine the contents of nuclides difficult to measure via correlation analyses is not new. Several earlier studies have been undertaken, of which the most comprehensive is presumably [NUR 85]. This study lists the results of several hundred single chemical analyses of nuclide vectors from various US American nuclear power plants. Additionally, the following correlations have been investigated:

with Co 60: C 14, Fe 55, Ni 59, Ni 63, Nb 94,

with Cs 137: Sr 90, Tc 99, I 129.

[NUR 85] provides a large database that is openly accessible and very valuable for comparisons. It may be, however, advisable to back up these data with more recent ones as the data are partly older than 10 years. More recently published papers like [WUR 90] which usually provide no actual data outline the procedure and describe the advantages of using scaling factors for the assay of nuclides that are difficult to measure.

The applicability of the correlation analysis method for the determination of difficult to measure nuclides has been sufficiently proven. Generally, there is also consensus about the choice of the key nuclides.



# **1.4.** Origin of the Data

From the beginning of this study, the main aim has been to include data from several countries, preferably recent measurements. Reasons for this can be:<sup>1</sup>

- The reactor operation has changed during the years. In general, the burn-up has been steadily increased. This causes different contents of fission products and transuranic nuclides in comparison to lower burn-up.
- The development of new decontamination procedures for the reactor loops and of the application of these procedures has helped to reduce the contents especially of strong  $\gamma$ -emitters in relation to other nuclides. Therefore, the ratio between corrosion products and Co 60 is influenced for correlation analyses.
- The use of data from various countries is interesting because comparisons of different reactor types and different kinds of reactor operation are possible.

Data from the following countries have been made available for this study:

1. Sweden:

various reports (see chapter 5.2., Swedish reports): [ABB 90], [CHE 92], [ING 93], [JOH 93], [SSI 84], [SKA 87], [TOV 86], [VAT 90], [VAT 91], [VAT 92], [VAT 93] (An overview of these studies can be found in appendix C.)

2. Germany:

Recent measurements from German energy producers, compiled under a sub-contract with the company "Wissenschaftlich-technische Ingenieurberatung GmbH" (WTI) [WTI 94a],

3. USA:

Data especially from the report [NUR 85], a few data also from [DOE 92],

4. Japan:

Data from the report [JAP 89].

The data are presented in different forms. For Germany, the compilation of nuclide specific measurements comprises complete nuclide vectors which usually also include Co 60 and Cs 137. From the Swedish reports, where several nuclide specific chemical analyses together with spectroscopic measurements have been reported, it has been possible to extract the data in a similar form. Unfortunately, Co 60 and Cs 137 not always have been measured.

For USA and Japan, data are available only as lists of data pairs of the difficult to measure nuclide and the corresponding key nuclide.

<sup>&</sup>lt;sup>1</sup>) A short discussion on how these points are reflected in the data analysis of this study can be found in chapter 4..



# 2. Water Chemistry and Reasons for Correlation between Nuclides

# 2.1. Water Chemistry in Light-Water Reactors

# 2.1.1. General Requirements for the Water

The water chemistry of light-water reactors including the activation processes in the core are important for the understanding of the processes that govern production and removal of radionuclides in the reactor water and therefore in the contamination.

The following requirements for the water in all light water reactors are generally applicable [FRE 84]:

- The probability for any type of corrosion should be as low as possible.
- The transfer of metal from the components into the water has to be minimized.
- Transport and deposition of crud has to be influenced in such a way, that the contamination of the primary circuit is as low as possible.
- Deposition of corrosion products especially on fuel elements should be avoided.

Interaction of the water with the commonly used materials in the primary circuits, like austenitic chromium nickel steels for components and pipes, Zircaloy for fuel cladding tubes, nickel chromium iron alloys like Inconel for steam generator pipes, and other alloys, result in certain specifications for the water chemistry. For example, certain values for the Li 7,  $H_2$ , Cl and  $O_2$  contents are required.

# 2.1.2. Special Requirements for the Boiling and Pressurized Water Reactors

With respect to the water chemistry, boiling water reactors and pressurized water reactors have slightly different requirements.

#### **Pressurized Water Reactor:**

The reactor water in a PWR usually fulfils the following specifications [FRE 84]:

Lithium-7-hydroxide	1 - 2 mg/kg
Hydrogen	2 - 4 mg/kg
Chloride	< 0.2 mg/kg
Oxygen	< 0.005 mg/kg
Silicic acid	< 4 mg/kg
Boric acid	< 0 - 2500 mg/kg
pH value at $T = 25 \text{ °C}$	< 5 - 10.5
Conductivity at $T = 25 $ °C	< 30 µS/cm

More recent experiences from PWR operation [MIL 94] show that it is advantageous to raising pH above 6.9 while retaining a long-term limit of 2.2 mg/kg lithium. For additional operational ad-



vantages, minimum hydrogen can be reduced from 25 to 15 cm<sup>3</sup>/kg (corresponding to a reduction from  $\approx 2.2$  mg/kg to  $\approx 1.4$  mg/kg). Zinc injection is also considered as a means to reduce penetration cracking and to control radiation fields.

These specifications serve to keep the metal transfer into the water as well as the deposition of corrosion products in the core region to a minimum. This in turn serves to keep the contamination as low as possible. The specification of chloride and oxygen is necessary as they are responsible for certain kinds of selective corrosion that has to be avoided.

Experience with different pH regimes have shown that an alkaline reactor operation is favourable whereas an acidic operation leads to an increase in the metal transfer into the reactor water. Alkaline operation also leads to an increase of solubility with increasing temperature which helps to shorten the time the dissolved metal needs to pass the hot core region (deposition is more likely in regions of lower temperature like the steam generators where no activation occurs). It also leads to less sorption and hence to a decrease of the deposition on surfaces.

An example for the chemical composition of undesirable deposits on fuel elements is given in [FRE 84] for the German nuclear power plant Obrigheim as follows:

At the lower section of the fuel elements:  $Fe_2O_3$ : 40 %, NiO: 25 %, SiO<sub>2</sub>, Cr<sub>2</sub>O<sub>3</sub>, ZrO<sub>2</sub> etc.

At the upper section of the fuel elements:  $Fe_2O_3$ : 20 %, NiO: 70 %, SiO<sub>2</sub>, Cr<sub>2</sub>O<sub>3</sub>, ZrO<sub>2</sub> etc.

The variability in the crud composition is mainly attributed to temperature differences. This, however, illustrates that the variability in the nuclide ratios can reach considerable values.

# **Boiling Water Reactor:**

The water chemistry in BWRs is different from PWRs. The main aims are of course similar to the PWR case, i.e. maintaining the water specifications, minimization of corrosion products, prevention of contamination etc., but as the reactor water is also transferred to the vapour phase and serves as the working medium for the turbines, differences in the chemical means to achieve these aims are necessary. [FRE 84]

BWRs are operated with neutral water. Due to the radiolysis at the fuel elements, the steam and all systems where the steam is condensed contain considerable amounts of oxygen. This leads to the choice of austenitic materials instead of ferritic steels preventing oxygen corrosion.

The reactor water in a BWR usually satisfies the following specifications [FRE 84]:

Chloride	< 0.2 mg/kg Cl
Silicic acid	<4 mg/kg SiO <sub>2</sub>
pH value at $T = 25 \text{ °C}$	6.5 - 8
Conductivity at $T = 25 \ ^{\circ}C$	< 1 µS/cm

More recent experiences from BWR operation summarized e.g. in [MIL 94] show that a higher hydrogen content protects core internals from stress corrosion cracking. In addition, zinc injection helps to reduce radiation fields.



### 2.1.3. Filtration

Filtration of the reactor water is a very important part of the strategy against contamination of a nuclear power plant. The coolant clean-up system mainly serves to maintain a water quality according to the specifications and to remove fission and corrosion products, but without changing the other water specifications like the pH value. The coolant clean-up system shall not be discussed in detail here. Only the nuclide specific efficiencies shall be outlined as this is directly related to the object of this study.

# **Pressurized Water Reactor:**

Mixed-bed filters together with mechanical filters are used for the reactor coolant clean-up system. The following table 2 lists nuclide specific decontamination factors that have been observed in the German nuclear power plant Stade (KKS): [FRE 84]

Nuclide	Decontami	nat	ion factor	Mean value
I 133	1000	-	10000	3000
I 135	500	-	3000	800
Cs 134/137	1	-	1.2	-
Cr 51	10	-	80	20
Mn 54	20	-	70	30
Co 58	10	-	100	20
Co 60	2	-	30	8

Table 2:Nuclide specific decontamination factors in PWRs [FRE 84]. The great variability for<br/>each nuclide and the nuclide specific differences should be noted.

A considerable variability of decontamination factors can be observed. Although table 2 does not list all nuclides that are of relevance in this study, it can be extrapolated that for other nuclides similar variabilities in the decontamination factors occur.

#### **Boiling Water Reactor:**

The reactor coolant clean-up system for BWRs differs from those of PWRs but of course serve the same purposes. A similar variability in decontamination factors can be expected.

More recent experiences from BWR operation [MIL 94] show that iron control in the feedwater system is essential to control radiation fields and deposits on fuel. Filtration of iron in the condensate system has been improved. This has been achieved by improved resins and new types of filters like hollow fibre filters.

Effects of filter efficiencies are also addressed in chapter 2.3.

# 2.2. Availability of Data and Correlation Analyses

The details of correlation analysis that is applied in this study are described in appendix A. In this chapter only the basic idea and the methods are described. Some considerations what kind of cor-



relation can be expected and which mechanisms have to be considered are presented in chapter 2.3.

# 2.2.1. Availability of Data

The data available in this study generally comprise either

- the full nuclide vectors of many nuclide specific chemical analyses which usually also give the abundance of Co 60 and/or Cs 137, or
- only data pairs of the difficult to measure nuclide and the corresponding key nuclide.

In the first case which is available for German and Swedish measurements, it is possible to perform correlations between every two nuclides for which measurements are available. For the second case as for the USA and Japan, only the data that have been pre-analyzed are available. That means, that the choice of a different key nuclide than the one already specified is not possible. It must be mentioned, however, that the choice of the key nuclides always is in accordance with the recommendations in other reports. - More complete information about how the data are available can be found in chapter 1.4. and in appendix C.

# 2.2.2. Methods for Correlation Analyses, Logarithmic Correlation

Details concerning correlation analyses have been included in appendix A. It is discussed in detail that a correlation between the logarithms of the data is the only appropriate way to assign equal weights to all data points as generally the data points cover a broad concentration range (several orders of magnitude).

In the correlation analyses shown in appendix D, the logarithm of the key nuclide concentration is generally plotted on the abscissa, while on the ordinate the logarithm of the concentration of the difficult to measure nuclide is shown.

From the equation of the fitted line which approximates the data points:  $y = a \cdot x + b$ , the dependence between the activities of the key nuclide and of the difficult to measure nuclide can be calculated:

$$A_{d} = a \cdot (A_{k})^{b} \tag{1}$$

with:

A<sub>d</sub>: Activity of difficult to measure nuclide,

 $A_k$ : Activity of the key nuclide,

a, b: coefficients from the correlation analysis.

If the same formation pathway, similar physical and chemical behaviour in the reactor water, similar filter efficiencies etc. applied to both the key nuclide and the difficult to measure nuclide, one would expect the exponent b in (1) to be unity or at least close to 1. This is discussed in more detail in chapter 2.3.

#### 2.2.3. Transformation between the Units of the Data

The preferable unit for the correlation analyses is Bq/g, but not all of the data used in this study are available in this unit. Most of the data are given as mass specific activity, i.e. Bq/g, Ci/Mg,



 $\mu$ Ci/g etc., but some are given as activity concentrations, i.e. Bq/cm<sup>3</sup> or similar units, and some as total activities, i.e. Bq.

In order to include these data points in a reasonable way, conversion factors have to be estimated. For the conversion of activity concentrations into mass specific activities, the density of the waste sample or the whole waste package has to assessed. The following transformations have been used:

A in  $Bq/g = 3.7 \cdot 10^4$  A in Ci/Mg =  $3.7 \cdot 10^4$  A in  $\mu$ Ci/g

A in Bq/g = A Bq/cm<sup>3</sup> /  $\rho$  in g/cm<sup>3</sup> = A in Bq/cm<sup>3</sup> / 2 g/cm<sup>3</sup>, where  $\rho$  is the waste density and 2 g/cm<sup>3</sup> can serve as a good estimate for  $\rho^2$ .

A in Bq/g = A in  $Bq / (V \text{ in } cm^3 \cdot \rho \text{ in } g/cm^3)$  where V is the volume of the sample or waste package ( $\rho$  see above).

It must be admitted that the uncertainties inherent in these transformations affect the position of a data point in the diagram and might therefore affect also slightly the slope of the correlation line. All the same, it must be emphasized that none of these transformations affects the ratio between the difficult to measure nuclide and the key nuclide. Therefore, this procedure which allows the inclusion of several data points which would otherwise have to be omitted has been applied. The transformation factors have been discussed with WTI [WTI 94b].

# 2.3. Reasons for Correlation between Radionuclides

In this chapter the reasons for a correlation between two nuclides are shortly discussed and then applied to a very simple model. Possible reasons for cases where the straightforward assumptions do not apply are discussed.

#### 2.3.1. Reasons to expect a correlation between radioactive isotopes

Radioactivity is generated in the reactor core. In the following it will be investigated to what degree a correlation between different isotopes can be expected and which factors disrupt a possible correlation. For this, simple first order equations to describe the generation of the radioactive isotopes will be used. In general two types of activity are generated in the core; firstly fission products escape from the fuel elements and secondly activation of elements in corrosion particles are transported through out the reactor and escape as contamination. The considerations here are aimed primarily at activation products but an analogy could be used to investigate fission products. The following nuclear reactions are examples of the type of activation products considered here:

$${}^{62}_{28}Ni + {}^{1}_{0}n \rightarrow {}^{63}_{28}Ni + \gamma$$
 (2)

2)

A value of  $\rho = 2 \text{ g/cm}^3$  is suitable for NPP wastes. The following average waste densities apply: compacted mixed waste:  $\approx 1.2 \text{ g/cm}^3$ , steel scrap 3.5-4 g/cm<sup>3</sup>, resins (loose):  $\approx 0.7 \text{ g/cm}^3$ , evaporator bottoms 1.5-1.8 g/cm<sup>3</sup>, cemented wastes: 1.8-2.0 g/cm<sup>3</sup>. Therefore, an average value of 1.8-2.0 g/cm<sup>3</sup> is reasonable. However, the choice of  $\rho$  has very little influence on the results of the correlation equations and parameters.



$${}^{58}_{28}Ni + {}^{1}_{0}n \to {}^{58}_{27}Co + {}^{1}_{1}p \tag{3}$$

$${}^{59}_{27}Co + {}^{1}_{0}n \to {}^{60}_{27}Co + \gamma \tag{4}$$

The possibility of correlations between different radioactive isotopes will be investigated with the help of the model presented in figure 1. This model reflects the activation process expected in a nuclear reactor. After the radioactive isotopes find their way out of the primary circuit they form part of the contamination found in the reactor and waste.



Figure 1 Model used to investigate correlation between different radioactive isotopes. Entrance (1) represents influx of source elements and exit (2) represents removal of the radioactive isotopes.

The schematic model in figure 1 is mathematically represented by equation (5),

$$\frac{dN}{dt} = \sigma M - \lambda N - \varepsilon N$$

$$N(t=0) = 0$$
(5)

where

- N : [moles] is the number of activated isotopes in the circuit,
- M : [moles] is the number of inactivated source elements,
- $\sigma$  : [1/s] is the activation rate constant,
- $\lambda$  : [1/s] is the decay constant and
- $\epsilon$  : [1/s] is the removal rate constant for the radioactive isotope from the circuit.

If it is assumed that the number of inactivated source elements remains relatively constant over the time period of interest then the solution to equation (5) is,



This assumption is reasonable considering that only a very small portion of the available elements in the circuit are activated and that the filtering system removes the corrosion as it is created leading to a long-term equilibrium in the system. Assuming that the decay constant is much smaller than the rate constant for removal of the activation from the system, this is the case for longer lived nuclides like Co 60 or Ni 63, and that the time of interest is long compared to the inverse of the removal rate constant, equation (6) is approximated to first order by equation (7).

$$N \approx \frac{\sigma}{\varepsilon} M$$

$$t \gg \frac{1}{\lambda} \gg \frac{1}{\varepsilon}$$
(7)

Each nuclide of interest has different properties and therefore  $\sigma$  and  $\varepsilon$  will in general be unique for each nuclide. Equation (8) is the general form of (7) where the different isotopes and source elements are indicated using the subscript j.

$$N_j = \frac{\sigma_j}{\varepsilon_j} M_j \tag{8}$$

If the ratio of two source elements is known,

$$\frac{M_2}{M_1} = \eta \tag{9}$$

then the relationship between their activation products is expressed as

$$N_2 = \frac{\sigma_2 \varepsilon_1}{\sigma_1 \varepsilon_2} \eta N_1 \tag{10}$$

Equation 10 shows that for a constant ratio between source elements a linear correlation between the product elements is expected. The experimental results show however that a linear correlation does not describe the data. Instead the data is described by

$$N_2 = b N_1^a \tag{11}$$

It can easily be shown that the exponent 'a' is not affected by radioactive decay. Including radioactive decay in the empirical equation (11) gives

$$N_{2} e^{-\lambda_{2} t} = b^{*} \left( N_{1} e^{-\lambda_{1} t} \right)^{a}$$
(12)



Using equation (12) to solve for 'b' in equation (11),

$$b = b^* e^{-(a\lambda_1 - \lambda_2) t}$$
<sup>(13)</sup>

shows that 'b' is affected by radioactive decay but that the power relationship is not influenced.

This does not exclude, however, that mixing data of 'old' and 'new' waste samples, i.e. samples of different age at the time of measurement, could indeed affect the slope of the correlation line, as is pointed out in figure 2. There it is assumed that two data sets are analyzed together, resulting in the solid correlation line. Each data set may consist of measurements of data of the same age. Therefore, separate correlation analyses of the single data sets result in correlation lines (dashed lines) that are parallel to each other, but deviate from the slope of the correlation line for the complete data.



log(Activity of key nuclide)

Figure 2: Example for effect of different 'ages' of data sets, i.e. time between generation of nuclides and measurement (arbitrary data). Two data sets are shown ( $\blacksquare$  and ●), each data set consists of measurements of the same 'age'. Correlation analysis for each data set results in parallel lines (similar slopes, dashed lines), whereas the correlation analysis for the complete data set (solid line) results in a different slope.

It must be emphasized, however, that the effect of different 'ages' of data sets is exaggerated in figure 2. In reality, it can be expected that data sets of different 'ages' would overlap more pronouncedly, i.e. cover about the same activity region, which would affect the slope of the overall correlation line only slightly.

Up to now only the amount of activity in the circuit has been discussed. Any number of mechanisms for removing activity from the circuit exist, for example a filter. These mechanisms are represented in the model schematic, figure 1, by the exit numbered 2. The amount of activity removed from the circuit is

$$C = \int_{0}^{\tau} \varepsilon N dt$$
 (14)



where C is the quantity removed. The quantity is converted to a concentration when divided by an appropriate volume. The relationship between quantity and activity is  $A = \lambda N$ .

If the removal rate is a constant then the amount of activity removed in a given time  $\tau$  is

$$C = \frac{\varepsilon \sigma M}{(\varepsilon + \lambda)^2} \left( \tau(\varepsilon + \lambda) - 1 + e^{-(\varepsilon + \lambda)\tau} \right)$$
(15)

Once again appropriate assumptions can be made to show the asymptotic behaviour of C for long (equation (16)) and short times (equation (17)).

$$C = \sigma M \tau$$

$$\varepsilon \gg \lambda \text{ and } \varepsilon \tau \gg 1$$

$$C = \sigma M \varepsilon \tau^{2}$$

$$\varepsilon \gg \lambda \text{ and } \varepsilon \tau \ll 1$$
(16)
(17)

The equations (16) and (17) show immediately that a simple removal model cannot account for the observed non-linear relationship between the activation products.

#### 2.3.2. Discussion

The analysis of the simple model shows that a linear correlation is expected for radioactive isotopes for which the source elements are linearly proportional. This should be the case e.g. for Co 58 and Ni 63 (see equations (2) and (3)) since the ratio of Ni 62 to Ni 58 should be independent of the reactor. The data for Co 58 / Ni 63 reflects the theoretical considerations, i.e. a = 1 (see chapter for Ni 63 in appendix D).

For Co 60 and Ni 63 (see equations (3) and (4)) this will only be true when the ratio of Co 59 to Ni 62 in the metals used are the same. The data shows that this correlation is not linear, i.e.  $a \approx 0.92$  (see chapter for Ni 63 in appendix D). The simple model used here cannot explain this observation but allows speculations as to the mechanism responsible for the observation. First, a varying ratio of the source elements in the metal used in the reactor could cause this since lower levels of Co 59 with the same levels of Ni 62 will on the average lead to the observed data. This hypothesis is testable by checking the relationship for each reactor. If the analysis for each reactor also shows the non-linear behaviour then a mechanism common to all reactors must be looked for. Using the model as a basis indicates that a non-linear effect or a higher order term (see equation (5)) is required. For example a weak coupling of the two activation rate constants, which could be caused by a change in the neutron absorption as an effect of the build up of one of the radioactive isotopes, would explain this effect. Another possible explanation is that the removable quantity in the circuit (see figure 1) depends on the quantity already removed (e.g. a loaded filter removes less activity). Then the removal rate constant  $\varepsilon$  in equation (5) depends on both radioactive isotopes which could lead to the observed data.

The effect seen here appears to be common to all reactors considered. This indicates that the nonlinear correlation between the isotopes is not an effect of a single filter type or reactor specific



chemistry but a common effect. It is important to test whether the effect is also observed in the data from a single waste stream from a single reactor. This is especially true since the non-linearity is not particularly strong for several correlations tested in this study (typically 0.9 < a < 1.1, but some stronger deviations from 1 exist). It is possible that the effect could occur from the averaging of the data from many waste streams and many reactors (also see [ING 93]). It should be noted that the more complete model used by ABB [ING 93] which is also based on linear first order differential equations also predicts that a = 1.

# 3. Measurement Techniques

Possibilities to measure the activities of the difficult to measure nuclides usually exist only after chemical decomposition of the waste matrix. Information about these analysis techniques together with the subsequent activity measurements have been listed for each of the nuclides in appendix D as available.

Additional information about basic measurement techniques and more elaborated methods have been compiled in appendix B. Direct measurement of difficult to measure nuclides that emit neutrons from spontaneous or induced fission events is to some extent possible with neutron measurements from the outside of waste packages. For the other nuclides, it seems that only correlation analyses can overcome the difficulties of assaying the nuclide contents of waste packages without the necessity of taking samples.

# 4. Nuclide Specific Results

In this chapter the results for the single nuclides as shown in appendix D are generalized. There the results of the analyses for the single nuclides, especially the scattering of the data points, the quality of the correlation etc. is presented.

Because of the different formation pathways, the correlations with the two key nuclides, Co 60 and Cs 137, are discussed separately.

# 4.1. General Results

In chapter 1.4. several possible reasons for performing nuclide specific correlation analyses have been addressed. Mainly, influences on the nuclide contents and the nuclide ratios can be expected because of changes in reactor operation with time or from country to country or because of the development of new decontamination procedures for the reactor loops to reduce the contents of strong  $\gamma$ -emitters. It is clear that such developments exist, but it is not possible to derive these changes from the correlation analyses of this study (Appendix D). In order to eliminate the influences of differences between certain plants, plant specific analyses of data would be necessary. However, this would require a significantly broader data base. In Germany, for example, the total number of full nuclide specific measurements would simply not be sufficient to allow the assess-



ment of changes with time or between single plants on a statistically significant basis. Some additional statements on country specific evaluations can be found in the nuclide specific chapters in Appendix D.

The waste samples that have been used for the analyses are taken from the reactor coolant, evaporator bottoms, solid waste, crud, filter cake, concentrates, resins, lubrication and coolant from the fuel pond (the complete list of samples can be found at the end of this report). Although for several nuclides samples are available only from some of these origins, the general situation can be characterized as follows: By including samples from a variety of origins it can be expected that the significance of the correlations is increased. The samples that were selected for the analyses are representative for the nuclide contents of waste packages.<sup>3</sup>

# 4.2. Correlation with Co 60

For the following nuclides, correlations with Co 60 (or with a nuclide that in turn has been correlated with Co 60) have been investigated:

• C 14, Cl 36, Ca 41, Ni 59 (with Ni 63), Ni 63, Mo 93, Pu 238

For C 14, according to [NUR 85] generic scaling factors should apply. It was, however, observed, that the scattering of data is quite large for BWR, and only for PWR a generic correlation seems possible.

For Ca 41, only very few measurements from concrete of German nuclear power plants are available. No correlation analysis was possible.

Ni 59 can be well correlated with Ni 63. An attempt to use a direct correlation of Ni 59 with Co 60 has failed.

Ni 63 is well correlated with Co 60.

Mo 93: no data are available.

Pu 238 can be correlated with Co 60. A better approach, however, seems to be to use correlation with Pu 239 + Pu 240, see below.

# 4.3. Correlation with Cs 137

For the following nuclides, a correlation with Cs 137 (or with a nuclide, that in turn has been correlated with Cs 137) has been investigated:

• Sr 90, Tc 99, I 129, Cs 135, U 234 (also with U), Np 237 (with U), Pu 238 (also with Pu 239), Pu 239/Pu 240, Pu 241 (also with Pu 239+Pu 240), Am 241

<sup>3)</sup> A method based on measurements of the activity of Co 60 and/or Cs 137 has been used in Germany already for several years for the determination of the nuclide specific total activity of waste containers [WUR 90]. Results of correlation analyses have been integrated in the German AVK (wasteflow, pursuit and product control system for wastes). The correlations used in these analyses are based on data that are similar to those presented in the present report.



The great number of data points together with the rather good quality of the correlation suggest that Sr 90 can be generically correlated with Cs 137.

For Tc 99 correlation analyses with Cs 137 are of medium quality. The behaviour of both nuclides in the clean-up system suggests plant specific correlation factors.

For I 129, correlations with Cs 137 have been investigated. Only data from Japan for BWR seem to be sufficiently correlated.

For the correlation of Cs 135 with Cs 137, no data points are available.

It has only been possible to investigate the correlation of Np 237 with U 235 and U 238. Too few data points were available to perform meaningful correlation analyses.

For the nuclides Pu 239 and Pu 240, for which the activities are usually not separated, the correlation with Co 60 provide better results than with Cs 137.

For the nuclides Am 241, Pu 238 and Pu 241 it has been found that a correlation with Cs 137, which could be expected to give good results, is not satisfactory. Instead, a correlation with Pu 239 + Pu 240 provides much better results, even the calculation of generic scaling factors seems possible. It can therefore be recommended to first perform a correlation analysis with Pu 239 + Pu 240 and then to use the previously established correlation of Pu 239 + Pu 240 with Cs 137, the latter perhaps on a plant specific basis.

# 4.4. Other nuclides

For tritium, no correlation with Co 60 or Cs 137 has been found. This applies both to BWR and PWR.

No actual measurements (only detection limits) have been reported for Cl 36. Therefore, a correlation analysis is not possible.

No measurements have been available for Mo 93.

Good correlations for U 234 have been found with U 235 or U 238. A correlation with Cs 137 is not possible. In addition, no correlation between U 235 or U 238 and Cs 137 exists. That implies that the knowledge of the correlations between U 234 and the other U isotopes cannot be used to calculate the correlation with Cs 137.

# 4.5. Overview of the Parameters of the Nuclide Specific Correlations

In this chapter, the results of the correlation analyses are graphically summarized. A more detailed discussion can be found in the nuclide specific chapters in appendix D.

# 4.5.1. Exponents

As can be seen from figure 3, for several nuclides, the exponents of the correlation analyses lie near 1, while for other nuclides the exponents deviate considerably from 1, both for PWR and BWR data. The strongest deviation show I 129 for PWR and Am 241 for BWR. All exponents have been taken from the analyses of the complete data sets (data from all countries). Additional



country specific plots would give different results (cf. nuclide specific chapters), but usually on a less significant statistical basis.

The graphs in figure 3 allow an easy comparison of the behaviour of the various nuclides. The error bars give a hint to the statistical significance of the correlations (a good correlation results in a smaller error range).



Figure 3: Exponents of the correlation analyses for PWR and BWR data.



4.5.2. Pre-Factor





From figure 4, the activity ratio of the difficult to measure nuclides and the key nuclides is visible. Differences between PWR and BWR data can easily be detected.



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# **Appendix A: Correlation Analysis**

#### A.1. Basic Method

The statistical analysis starts with the set of measured data from n separate measurements, i.e. n data pairs that are denoted as  $(x_i, y_i)$ , i=1,..,n.

The data pairs are usually visualised in a x-y scatter chart and form a cloud of points that are concentrated around a straight line if the data are correlated. This so-called regression line is described by the equation

$$\hat{y} = a + b \cdot x \tag{1}$$

where x is given and 3 is the computed value for the given s (the ^ serves to mark computed in contrast to measured values). The parameters a and b are given by the equations

$$b = \frac{\sum_{i} (x_{i} - \overline{x}) \cdot (y_{i} - \overline{y})}{\sum_{i} (x_{i} - \overline{x})^{2}} = \frac{\sum_{i} x_{i} \cdot y_{i} - \frac{1}{n} \sum_{i} x_{i} \sum_{i} y_{i}}{\sum_{i} x_{i}^{2} - \frac{1}{n} \left(\sum_{i} x_{i}\right)^{2}}$$

$$a = \frac{\sum_{i} y_{i} - b \cdot \sum_{i} x_{i}}{n}$$
(2)

in the usual way. The correlation coefficient r is given by

$$r = \frac{\sum_{i} x_{i} y_{i} - \frac{1}{n} \left( \sum_{i} x_{i} \right) \left( \sum_{i} y_{i} \right)}{\sqrt{\left[ \sum_{i} x_{i}^{2} - \frac{1}{n} \left( \sum_{i} x_{i} \right)^{2} \right] \cdot \left[ \sum_{i} y_{i}^{2} - \frac{1}{n} \left( \sum_{i} y_{i} \right)^{2} \right]}}$$
(3)

The correlation coefficient expresses the strength of the correlation: if r = 0, no correlation exists, if  $r = \pm 1$ , the correlation is perfect. For a given set of measured nuclide concentrations, the correlation coefficient will usually be 0 < r < 1.

# A.2. Determination of the Region of Confidence

A value of the correlation coefficient r near  $\pm 1$  is not enough to ensure that the correlation itself is meaningful as r can only be used to assess the quality of a correlation if the following conditions are met (conditions 1 and 4 are most important):

- 1. linear connection between x and y
- 2. continuity of x and y
- 3. normal distribution for x and y



4. independence of each data pair

In order to test these conditions, various statistical tests have been developed. An important test criterion is the value of the F distribution for two given data sets.

# A.2.1. F distribution

The F distribution serves for the comparison between two variances of data and also in the correlation analysis. The variable

$$F = \frac{S_1^2}{S_2^2} \tag{4}$$

(with S<sub>1</sub>, S<sub>2</sub>: variances of 2 random samples of size n<sub>1</sub> and n<sub>2</sub>, respectively, taken from 2 normally distributed populations)

follows a *F* distribution with the parameters  $v_1 = n_1 - 1$  and  $v_2 = n_2 - 1$ . The F distribution is a continuous asymmetrical distribution from 0 to  $\infty$ . The probability distribution of F is shown in figure 1. It is L shaped for  $v_1 \le 2$  and has one maximum for  $v_1 > 2$ .



Figure 1 Probability density of the F distribution for the two cases  $F(v_1=1, v_2=5)$  and  $F(v_1=10, v_2=10)$ 

For each given set of values  $v_1$  and  $v_2$  a value of the safety limit of the F distribution can be computed for a given probability  $\alpha$ . For the correlation analysis the parameters  $v_1$  and  $v_2$  take the role of degrees of freedom:

- $v_1 = 2 = \text{const.}$ , as 2 coefficients have to be computed for the equation of the regression line (equation (2)), i.e. only 2 degrees of freedom, and
- $v_2 = n-2$ , (n: total number of data points), n minus 2 degrees of freedom because 2 data points are at least necessary for the calculation of the 2 coefficients in equation (2).



The following table 1 shows some values of the F distribution for  $v_1 = 1$  and 2 and the two probabilities  $\alpha = 5 \%$  and  $\alpha = 2.5 \%$ . These two probabilities are chosen as the region of confidence is usually calculated for 95 % ( $\alpha = 5 \%$ ) and the two-sided and one-sided F distributions are connected according to

$$F_{v_1,v_2,\alpha}^{two-sided} = F_{v_1,v_2,\alpha/2}^{one-sided}$$
(5)

Table 1	Upper significance	points for the F	distribution for $v_1 = 1$	and 2 and $\alpha = 5\%$ and 2	5 %
---------	--------------------	------------------	----------------------------	--------------------------------	-----

2.5%	2.5%	5.0%	5.0%		ά
2	1	2	1	ĩ	v
799	648	199	161		ν <sub>2</sub> 1
39.0	38.5	19.0	18.5		2
16.0	17.4	9.6	10.1		3
10.7	12.2	6.94	7.71		4
8.43	10.0	5.79	6.61		
7.26	8.81	5.14	5.99		~6°
6.06	7.57	4.46	5.32		8
5.46	6.94	4.10	4.96		10
5.10	6.55	3.89	4.75		12
4.86	6.30	3.74	4.60		14
4.69	6.12	3.63	4.49		16
4.56	5.98	3.55	4.41		18
4.46	5.87	3.49	4.35		20
4 <b>.2</b> 9	5.69	3.39	4.24		25
4.18	5.57	3.32	4.17		30
4.05	5.42	3.23	4.08		40
3.93	5.29	3.15	4.00		÷60
3.80	5.15	3.07	3.92		120
3.69	5.02	3.00	3.84		8



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# A.2.2. Construction of the Region of Confidence

The region of confidence around a given regression line is centred around the regression line and limited by an upper and a lower hyperbolic curve as shown in figure 2. The following different regions of confidence exist:

- 1. Region of confidence for the entire regression line,
- 2. Region of confidence for the expectation value Y at X = x,
- 3. Prediction interval for a value Y at X = x.

In this study, the third kind of region of confidence is of interest, because for a given value of the activity of the key nuclide, a prediction of the activity of the correlated difficult to measure nuclide is intended. The calculation of this region of confidence is described below.



Figure 2: Region of confidence (dashed lines) around regression line (solid) (arbitrary data points)

The role of the F distribution in the calculation of the region of confidence for the regression line is expressed in the following equations from which the upper and lower boundary values  $\hat{y}_x^{\pm}$  for the region of confidence for any given x can be calculated:

1. The mean value  $\overline{x}$  of the x values of all data pairs is calculated as

$$\overline{x} = \frac{1}{n} \sum_{i=1}^{n} x_i \tag{6}$$

2. The sum  $Q_x$  of the squares of the deviations of the x values from  $\bar{x}$  is calculated as:

$$Q_x = \sum_i \left(x_i - \overline{x}\right)^2 \tag{7}$$

3. The residual variance  $s_{y,x}$  is calculated from the y values of the data pairs  $(y_i)$  and the corresponding value  $\hat{y}$  of the regression line (equation (2)):



$$s_{y \cdot x} = \sqrt{\frac{\sum_{i} (y_{i} - \hat{y})^{2}}{n - 2}}$$
(8)

- 4. The value of the F distribution for  $v_2 = n-2$  ( $v_1 = 1$ ,  $\alpha = 2.5$  %),  $F_{(1;n-2;0.025)}$ , is taken from tables.
- 5. The width  $B_x$  of the region of confidence for a given x is then calculated as

$$B_{x} = \sqrt{F_{(1,n-2,\alpha)}} \cdot s_{y \cdot x} \cdot \sqrt{1 + \frac{1}{n} + \frac{(x - \overline{x})^{2}}{Q_{x}}}$$
(9)

6. At last the y values of the upper and lower boundary of the region of confidence,  $\hat{y}_x^{\pm}$ , are calculated as

$$\hat{y}_x^{\pm} = \hat{y}_x \pm B_x \tag{10}$$

For the graphical representation, a sufficiently large number of evenly spaced values  $\hat{x}$  between  $x_{\min}$  and  $x_{\max}$  (where  $x_{\min}$  and  $x_{\max}$  are the lower and upper value of the x values of the given data pairs) is chosen, the corresponding values  $\hat{y}_x^{\pm}$  are computed and the data points  $(\hat{x}, \hat{y}_x^{\pm})$  are connected by a smooth line. These hyperbolic curves are centred around the regression line.

# A.3 Correlation Analysis

This chapter discusses whether a correlation between the logarithms of the activity values (loglog) or between the original activities (lin-lin) should be performed. As an example, both possibilities are compared for Ag 110m.



×	ÿ	
1999 B. C.		
1,20E-06	1,56E-06	
5,40E-06	7,08E-07	
9,00E-06	4,90E-07	
1,40E-05	6,80E-07	
1,67E-05	8,70E-07	
4,02E-05	2,20E-06	
7,90E-05	3,80E-07	
1,31E-04	7,40E-06	
1,38E-04	2,72E-06	
1,77E-04	8,86E-06	
2,96E-04	7,43E-05	
8,10E-04	1,30E-04	
3,32E-03	1,45E-05	
1,00E-02	3,40E-04	
1,56E-02	3,29E-04	
2,81E-02	3,13E-04	
3,87E-02	5,70E-04	
4,60E-02	1,20E-04	
6,11E-02	4,75E-04	
8,53E-02	1,86E-03	
1,55E-01	5,99E-03	
2,41E-01	6,89E-03	
3,79E-01	2,15E-03	
7,80E-01	1,90E-02	
9,54E-01	7,28E-03	
1,10E+00	2,48E-01	
5,94E+00	9,97E-02	
1,64E+01	1,19E-01	
2,90E+01	1,93E-01	
6,52E+01	1,90E-01	
2.12E+02	5.60E-01	

Interval boundaries	Frequency of x- values in interval	Frequency of y- values in interval		
1E-07	0	0		
1E-06	0	5		
1E-05	3	5		
1E-04	4	2		
1E-03	5	7		
1E-02	2	5		
1E-01	6	2		
1E+00	5	5		
1E+01	2	0		
1E+02	3	0		
1L+03		0		
[				
frequency of data points trequency of data points 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2				
1E-07 1E-06 1E-06	장 않 않 다 보 보 보 Interval	1E+00 1E+01 1E+02 1E+03		

Fig. 3: Arbitrary data set (x and y values, sorted by the x values), frequency of x and y data in the decades from  $10^{-7}$  to  $10^3$  and graph of this frequency distribution

#### A.3.1. General Considerations

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The activity data available for the present study are distributed over several orders of magnitude. When investigating the original data it is commonly found that in each decade between the minimum and the maximum value there are several data points (depending on the nuclide and on the waste stream the samples were taken from). The following arbitrary worksheet serves to illustrate this (figure 3).

It can be seen that in each decade several data points are present and that the frequency of data points in each decade varies only slightly. Each data point is assumed to have the same accuracy and statistical weight. That means that for a correlation analysis each data point should have the same significance.



It is most easily seen how a lin-lin or a log-log analysis weighs the data points if the data are plotted in a lin-lin or log-log scaled diagram as in figure 4. On the left hand side (figure 4a), a plot with linear x and y axes is displayed for the data points shown in figure 3. Only the points with x and y values above approximately 0.1 and 0.01, respectively, can be distinguished at all. On the right hand side (figure 4b), the same data points are displayed in a logarithmically scaled diagram, which is equivalent to plotting the logarithms of the logarithms of the original data in a linearlinear plot. The strong correlation of the *logarithms* can be clearly seen. Therefore it is clear that a logarithmically scaled diagram is the only way to display data points of the kind dealt with in the present study.

Coming back to the question of the best way to perform a correlation analysis, in the lin-lin case (figure 4a) the few data points with high values<sup>4</sup> would influence the correlation very strongly while the many data points with low values influence the correlation only slightly. If, however, a correlation is performed for the log-log case (figure 4b), all data points will contribute with the same statistical weight. This question is discussed further in the next section.



Figure 4: Comparison of a) linearly and b) logarithmically scaled plots of the data shown in fig. 3

# A.3.2 Comparison of the two correlation methods for Ag 110m

The influence of performing a correlation between the original data (linear-linear plot) or between the logarithms of the data (log-log plot) is illustrated for the example of Ag 110m. Data for the US nuclear power plants (PWR) are used.

Figure 5 shows a correlation between the logarithms of the activities of Ag 110m and Co 60. The statistical evaluation shows that the correlation is of good quality (r=0.97) and meaningful (F=931). The slope is nearly 1 (a=0.92) indicating that the formation Ag 110m and Co 60 is accomplished in the same or very similar ways. The good quality of the correlation is true for the entire range of data ( $10^{-4}$  to  $10^{7}$  Bq/g Co 60).

<sup>4)</sup> As all measurements values must be zero or positive (activities!), there is no need to speak of the *absolute* values.




Figure 5: Data points and correlation between the logarithms of the activities of Ag 110m and Co 60 for PWRs, USA. The correlation has been performed on the logarithms of the data.



Figure 6: Data points and correlation between the activities of Ag 110m and Co 60 for PWRs, USA. The correlation has been performed on the original data.

Figure 6 shows the same data in a linearly scaled plot with a correlation performed on the original data (not on the logarithms of the data). It can be seen that the quality of the correlation is worse than in figure 5, as the correlation coefficient is smaller (r=0.66). Figure 7a shows that the correlation on the linear data from figure 6 deviates strongly from the actual data in the lower range. However, for higher values both correlations give similar results (enlarged diagram in figure 7b).





Figure 7: Data points and correlations between the activities of Ag 110m and Co 60 (for PWRs, USA). The correlations are performed on the logarithms of the data (log-log case, fig. 5) and on actual data (linlin case, fig. 6), respectively. Diagram a shows the entire range indicating that the two correlations deviate strongly for small values. Diagram b shows only the rightmost part of data points indicating that the two correlations are nearly identical for higher values. The correlations have been taken from fig. 5 (loglog) and fig. 6 (lin-lin).

### A.3.3. Conclusions

As a result from the previous discussions, it can be deduced that a correlation between the logarithms of the activities usually will give the best results. Therefore, for all nuclides investigated in this study, the first approach will be a correlation between the logarithms.

At the moment it is unclear whether there is any alternative to a correlation with a polynomial of degree 1 (i.e. a function y = ax + b) between the logarithms of the measured data points. A possi-



ble alternative would be a correlation with a polynom of higher degree (e.g.  $y = ax^2 + bx + c$ ) between the original data points. However, the chemical or physical formation or absorption pathways (e.g. different efficiencies of ion exchange resins) of the nuclides being considered do not suggest such an approach.



# Appendix B: Measurement Methods

Some important detectors and basic measurement methods for the determination of radioactive isotopes are listed in the first part of this paragraph (for a complete overview, see e.g. [NCR 85]). In the second part, additional methods that allow determination of difficult to measure nuclides non-destructively are presented.

### **B.1. Destructive Methods**

This paragraph gives a short overview of destructive measurement methods, i.e. methods that require samples being taken from the waste package.

### **B.1.1** Ionization chambers

Ionization chambers consist of a volume of gas across which an electric field is applied. It may be in cylindrical form with a cylindrical cathode and an axial insulated rod anode. The measurement process is based on the production of positive ions and electrons in the gas produced by the radiation to be measured. Electrons and ions move to the electrodes under the influence of the electric field resulting in a current. This ionization current that is proportional to the number of ion pairs produced per unit time can then be measured. A single incident particle depositing its energy in the gas of the ionization chamber will give rise to a number of ion pairs proportional to the deposited energy. An externally applied voltage will cause a pulse of current to flow between anode and cathode of the chamber. A chamber operated in this mode is referred to as a pulse ionization chamber.

The use of a pulse ionization chamber at low particle energies is limited by the inherent noise of the associated amplifier. This noise is typically equivalent to the arrival of 200 electrons which might represent an energy of about 7 keV dissipated by a particle traversing the chamber. [NCR 85]

### **B.1.2 Proportional Counters**

When the electric field strength at the centre electrode of a pulse ionization chamber is increased above a certain level, the size of the output pulse starts to increase but is still proportional to the initial ionization. A device operated in this mode is call a proportional counter. It has the great advantage of allowing to detect a very low initial ionization. It can be used to determine the energy of particles and photons if they are totally absorbed in the counting gas, or to particle counting without energy discrimination. A proportional counter is suitable for determination of low-energy X and gamma radiation. With the use of low-noise amplifiers counting of particles with energies down to a few keV is possible.



### **B.1.3** Scintillation Detectors

Substances that emit visible light when exposed to nuclear radiation are suited for scintillation detectors. These detectors consist of the scintillator, which emits light and which is surrounded by a thin reflector of aluminium oxide to increase the collection efficiency of the emitted light. The photons produced in the scintillator reach the cathode surface of an attached electron-multiplier phototube where they are converted into electrons by the photoelectric process. In the phototube, the minute photocurrent is amplified to a level that can be suitably handled in conventional electronic amplifiers. The output pulse can be fed to a multichannel pulse-height analyzer so that the energy spectrum of the radiation being measured can be recorded.

A very important scintillation medium is thallium-activated sodium-iodide crystal, NaI(Tl), for the measurement of radiation coming from media outside the detector. However, it is also possible to incorporate the nuclides to be measured like low-energy beta-emitters (H 3, C 14) into a scintillating liquid. These are generally composed of one or more fluorescent solutes in an organic solvent. The radioactive matter is introduced into, and mixed with, the liquid scintillator. The radiation energy ionizes and excitates the solvent. This energy is transferred to the solute and re-emitted as photons.

Liquid-scintillation counting is useful in making relative measurements of beta-particle emitters with energies above several keV, and direct measurements of alpha-particle emitters. This method is e.g. frequently used for the direct assay of high-energy beta emitters such as P 32 and Y 90, but also for low-energy beta-emitting radionuclides such as H 3, C 14, S 35 and Ni 63, and electron-capture radionuclides such as Fe 55 and I 125.

Before a measurement of alpha-, beta- or X-ray emitting nuclides is possible, these nuclides generally have to be separated from the mixture of other nuclides or from an absorbing matrix in which they are embedded. Several chemical separation techniques exist some of which are described for the various nuclides in the respective chapters in appendix D.

In most instances, counting samples are prepared by pipetting the scintillator formulation into the vial and then adding a known amount of an aqueous solution of the sample. Other methods are the suspension of scintillators into the aqueous radioactive solution to form a two-phase counting system, the addition of gel scintillators for insoluble material or solutions with high solids contents, or the counting with solid-source supports like membranes, filters etc.

#### B.1.4 Semiconductor Detectors

Radiation detectors on the basis of semiconductors have similar high detection efficiency as scintillation detectors, but have much better energy resolution. Pulse-amplitude analyzing systems employing detectors fabricated from single crystals of Si and Ge are commonly used for radionuclide analysis.

If a voltage is applied across the detector crystal, electrons that have been raised to the conduction bands of the semiconductor by excitation, and the holes in the valence band are free to move to the electrodes where they are collected. Various crystals are suitable for semiconductor detectors. Crystals with residual impurities (as in the early period of development) can be compensated by drifting Li ions through large volumes of the crystal and thus creating a p-i-n (positive-intrinsicnegative) structure. A disadvantage of a drifted crystal is that it has to be kept at cryogenic temperatures throughout its lifetime as the mobility of Li ions is not negligible at room temperatures.



Semiconductor crystals free from impurities or structural defects can be used directly for the fabrication of detectors.

### B.1.5 Gamma Spectroscopy

Gamma spectroscopy measures the electromagnetic radiation that is emitted from  $\alpha$  or  $\beta$  decay of most nuclides. Usually semiconductor detectors, mostly Ge(Li) detectors, together with multichannel analyzers, are used for the measurement. Analysis of nuclide mixtures is possible without sample preparation or extraction of single elements. Only for the measurement of certain lowenergy  $\gamma$ -emitters, like I 129, additional preparation steps are necessary.

### B.1.6 Measurement of Alpha-Particle Emitting Nuclides

Because of the very short ranges of alpha particles (less than 0.1 mm in liquid and solid samples), the amount of matter between the radioactive source and the alpha-particle detector must be minimized, e.g. by close proximity, placement of source and detector in a vacuum chamber or mounting the source inside the detector. While for gross alpha-particle measurements the source is usually at least as thick as the range of the alpha-particles in order to attain maximum sensitivity, for alpha-spectroscopy the source usually is in a very thin layer to provide maximum resolution. [NCR 85]

Scintillation counters, gas-filled detectors and alpha-particle spectrometers (mainly pulseionization chambers and semiconductor detectors) can be used for measuring. Spectrometers and signal processing resemble those of gamma spectrometry. Problems may arise from overlapping peaks because of only small energy differences of the alpha decays. For example, the peaks of Pu 239 and Pu 240 are so close that only the sum can be detected. Other important examples for peak overlapping are Pu 238 and Am 241.

### B.1.7 Measurement of Beta-Particle Emitting Nuclides

Beta-particle emitting nuclides like H 3, C 14 or Sr 90 are usually measured with liquid scintillation detectors. Concerning the determination of radioactive inventories of wastes, the measurement of tritium with scintillation detectors is quite common.

Because of the continuous nature of a beta-particle energy spectrum, spectrometric identification and quantification of a sample containing several beta emitters are generally difficult and usually require radiochemical separations [NCR 85].

### B.2 Non-destructive Methods

This paragraph describes some methods for the non-destructive detection of nuclides, i.e. methods for which no sample has to be taken from the waste. Examples for such techniques are the following:

- Neutron balance measurements (Pu detection),
- Neutron interrogation (determination of fissile U contents),
- Computer tomography of waste packages with active and passive gamma measurements,



• Gamma spectrometric measurements on waste packages.

Aside from cases where higher concentrations of U or Pu are present in the waste packages, these measurement methods do not allow detection of the difficult to measure nuclides investigated in this study.

### B.2.1 Neutron Balance Measurements for Pu Detection

Methods for measuring the concentration of Pu within a mixture of other nuclides nondestructively already exist. The lowest sensitivity is in the region of several mg Pu in the whole waste package. Pu isotopes are alpha, beta and gamma active, fission spontaneously releasing neutrons and are fissioned by neutrons. Only neutrons have a sufficiently long mean free path to escape from the items and to be measured. Their intensity can be used for the determination of the Pu contents in a non-destructive assay instrument. The neutrons originate from spontaneous fissions,  $(\alpha,n)$  reactions with light nuclei and multiplication of primary neutrons. Only the spontaneous fission events are directly proportional to the mass of Pu and must be separated from the other two sources of neutrons. This can be achieved by certain statistical properties that only apply to this kind of neutron emission. [KLE 88], [KLE 91], [CIF 86].

The Pu isotopes Pu 238, Pu 240 and Pu 242 undergo spontaneous fission releasing neutrons. In addition the Pu alpha radiation leads to neutrons from  $(\alpha,n)$  reactions with the heavy isotopes of O and F [KÜC 88]. The energies of the neutrons emitted from these processes are not specific for the Pu isotopes, but the total amount of Pu can be calculated from the neutron intensity if the isotopic composition of the Pu is known. For increasing the sensitivity the neutrons are thermalized and counted with BF<sub>3</sub> or H 3 counters that are mounted around the waste drum. The detection limit can be as low as 10 mg Pu for a 200 l-drum. The measurement can be influenced by the presence of Cm.

In addition, under certain conditions the characteristic gamma radiation can be used for an evaluation of the Pu contents. The waste matrix has to be sufficiently transparent for the gamma radiation and the total gamma activity of other isotopes has to be low enough [KÜC 88]. For example, in a 200 l-drum the quantitative determination of the Pu contents is only possible if the matrix density is less than 0.3-0.5 g/cm<sup>3</sup> which is usually the case only for certain combustible wastes. A concrete matrix or high scrap contents in the waste provide a shielding that is not penetrated by the gamma radiation. Therefore, this technique is mostly used for wastes from fuel element fabrication that does not contain gamma sources interfering with the gamma lines of Pu.

A schematic view of a device working on the basis of neutron detection is shown in the next figure [KLE 91]. The sample is surrounded by piles of neutron detectors. Three neutron counters embedded in polyethylene that serves as a moderator are grouped to one unit. The whole unit is shielded with Cd that permits only the fast neutrons to pass. After the thermalization in the polyethylene the neutrons that are not counted are absorbed by the Cd. In that way backscattering is avoided. The gamma underground signal is reduced by a steel plate shielding.

The block data sampling includes a time correlation analyzer (TCA). In the next figure the main units of the TCA are drawn. The pulse train generator converts signals from the detectors that enter the 16 channels of the unit at the same time into sequential signals (pulse train). The units for the pulse fluctuation analysis and the pulse-to-pulse analysis work in parallel to analyze this pulse train. Both utilize a different approach for grouping the incoming neutron signals according to the



time and to detect coincidences. With cross and auto correlation methods a discrimination between neutrons from  $(\alpha,n)$ -reactions (uncorrelated single events) and from spontaneous fissions (correlated ensembles of events) is possible. From the knowledge of spontaneous fission events the Pu fraction can be calculated.



Figure 8: Principle of an automated measuring system for the non-destructive analysis of radioactive waste packages [KLE 91]

### **B.2.2** Neutron interrogation for Determination of Fissile U Contents

For the determination of fissile U in closed waste barrels or storage containers for irradiated fuel the active neutron interrogation principle can be used. The waste packages are irradiated with neutrons from Sb-Be sources and the prompt neutrons from fission events of nuclides in the waste package are measured. The principle of this technique is shown in the following figure [FIL 85]. The neutron source consists of the gamma emitter Sb 124 that induces neutrons from a Be block; removal of the Sb 124 stops the neutron emission. The following  $(\gamma,n)$  process is utilized:

 ${}^{9}_{4}\text{Be} + {}^{0}_{0}\gamma \rightarrow {}^{8}_{4}\text{B} + {}^{1}_{0}n$ 

The waste package is continuously rotated during the measurement in order to average over the whole volume. The neutrons are detected in  $BF_3$  counters after thermalization in polyethylene.



Figure 9: Principle of measuring fissile U in storage containers for irradiated fuel by interrogation with neutrons [FIL 85]

The irradiation with neutrons of low energy induces fissions in the fissile isotopes generating prompt neutrons of high energy. Although all fissile nuclides (e.g. U 233, U 235, Pu 239, Pu 241) contribute to these neutrons the knowledge of the origin of the waste package helps to identify the nuclides that really are contained in the sample. For example, the only relevant nuclide contained in enriched, natural and depleted uranium that can be fissioned by slow neutrons is U 235 (irradiated samples from the Th fuel cycle [i.e. from high-temperature reactors] contain U 235, U 233 and only traces of Pu and so on).

With the apparatus discussed in [FIL 85] it is possible to measure U contents in the range from 1 g up to 500 g in waste packages of a total volume of 220 l with surface dose rates of up to 2 Sv/h. The upper boundary originates from the pile-up of  $\gamma$  pulses into the region of the neutron pulses which is due to the high dose rate. A change of the detectors could still enlarge the measurement range.

A more recent application of active neutron interrogation (together with passive high-resolution gamma measurements in conjunction with waste form evaluations and Monte Carlo modelling studies) is described in [BEC 94]. The waste to be investigated normally contains weapons grade Pu, depleted U and Am 241 in varying composition (fractions can vary over several orders of magnitude, in addition, the composition of depleted U can vary). The non-destructive measurement process is designed to account for the presence and variation of these radionuclides and compounds. The active mode response of the drum neutron counter responds to waste entrained fissile materials such as Pu 239, Pu 241, U 235, U 233 etc., but the technique of thermal neutron interrogation cannot distinguish between two or more fissile species nor can it identify and quantify Am 241. Therefore, neutron interrogation is supplemented with high resolution gamma spectroscopy to identify those radionuclides present in the solidified waste drum and establish the mass of each relative to the Pu 239 isotope.<sup>5</sup>

5)

The procedure for deriving the activities of the other nuclides with high resolution gamma spectroscopy, waste stream analysis and calculations is described in detail in [BEC 94] and is not repeated here.



### B.2.3 Computer Tomography of Waste Packages with Active and Passive Gamma Measurements

A way for achieving a rather detailed overview over the contents of waste packages without penetration is the computer tomography of the gamma activity [DUW 87]. Depending on the configuration of the apparatus either the density or the activity distribution can be measured. In the transmission configuration the radiation from an external source (typically 10 Ci) is detected through the waste container and the weakening of the gamma beam serves as a measure for the density. In emission the radiation from the waste itself is measured. The activity distribution in the plane is computed from the radiation measurement together with the information about the density and is recorded for one horizontal plane at one time giving a two-dimensional distribution. Threedimensional distributions therefore take considerable time to be measured. In the original version of this device the gamma radiation of Cs 134 and Cs 137 in the region between 600 and 800 keV was detected. In principle - especially for the emission measurement set-up - it is possible to detect the spatial distribution of various nuclides separately.

A schematic view of this device is shown in the next figure. A computer controls the angular and lateral position of the waste package mounted on a turntable and computes an image of the measured plane after one or more revolutions from the measured gamma activity.

This technique is still under development. The spatial resolution, the time required for one measurement and the limits for the detection of single nuclides depend strongly on the detector, the measuring time, the statistics, the peak detecting software etc.



Figure 10: Principle of a measuring device for computer tomography with gamma spectroscopy on intermediate level waste packages [DUW 87]

More recent developments have been presented for example in [SIM 95]. A waste drum measuring facility (GME) was developed for completely characterizing waste drums for final disposal in Switzerland. The system determines the mass of the waste drum and the dose rate at 1 m distance and performs wipe tests on the surface of the drum in order to discover possible contaminations. The drum is turned with 1 rotation per minute. The measurement itself is performed by a gamma



spectrometer with a very pure Ge detector, a collimator system and nuclide software. Gamma emitters are identified and their source strength stated<sup>6</sup>. The performance of the system is completely automatic aside from the loading/unloading operations.

A different approach towards spatial resolution of the activity distribution is to use several detectors simultaneously both for the vertical and horizontal resolution. A measurement device that uses two sets of detectors (each with 12 NaI(Tl) detectors) to obtain two-dimensional activity distributions both for horizontal and vertical planes [YOS 94] with a spatial resolution in the range of 5 to 10 cm and a detection limit of 0.4 Bq/g. The apparatus was designed to measure low-level decommissioning waste for recycling/reuse with a throughput of 500 kg/h. An application for measurement of waste packages for final repositories would require certain changes, but it can be seen that the use of several parallel detectors can increase the throughput quite effectively compared with the use of a single detector.

In some cases, waste drum measuring facilities are complemented with radiography and tomography in order to gain a picture of the mass and density distribution inside a waste package which serves a better interpretation of the results of the gamma measurements [RON 94].

### B.2.4 Gamma Spectrometric Measurements on Waste Packages

Gamma spectrometric measurements are performed on waste packages for characterizing waste drums for final disposal. An example has been given in [SIM 95] and is described above. Further developments are presented in [DIO 94]: A measuring device for the radiological characterization of supercompacted low-level wastes uses a segmented gamma spectroscopy system suitable for the identification and activity determination of the main radionuclides in 220 l waste packages or in 320 l overpacks. A hyperpure Ge detector is used for the measurement of the most representative radionuclides Co 60 and Cs 137 and for Mn 54, Zn 65 and Cs 134. Sensitivities for these nuclides are in the range from 0.3 to 1.5 Bq/g, depending on density, matrix, and package volume. Comparisons between known activities of calibration sources and measurements show deviations of about 10 %. Appropriate calibration factors for various geometries have been established.

### B.2.5 Correlation Analyses and "Fingerprinting"

If none of the above methods is applicable, then the only possibility is to use correlation analyses. For example, the data presented in this study can serve as a generic basis for wastes from nuclear power plants with light-water reactors.

Because many reports have already been published on the subject of correlation analyses, only two research works shall be mentioned here in more detail. The work presented in [FUD 92] and [MCM 94] concentrated on wastes from decommissioning projects in the UK. "Fingerprints" of some relevant waste streams have been established prior to dismantling by performing comprehensive radionuclide analyses of small but statistically significant numbers of samples coupled with a widespread radioactive survey of the waste to assess its overall activity. The activities of some low energy beta-emitting and electron capture nuclides and other difficult to measure nu-

<sup>6)</sup> 

No information is given in [SIM 95] on scaling factors or correlations by which difficult to measure nuclides are evaluated from the measurement of the gamma emitters.



7)

clides (H 3, Ca 14, Ca 41, Fe 55, Ni 59, Ni 63, Fe 55, U) have been measured, and appropriate ratios to gamma-emitting nuclides have been established. It has been found that the results are well applicable for activated wastes. For contaminated wastes, however, activities predicted from the "fingerprints" were found to deviate up to an order of magnitude from the measured values because this type of waste is often highly heterogeneous.<sup>7</sup>

A second example for using correlations and uncertainties for the determination of nuclide contents in low-level solid waste packages is presented in [GED 95]. At Hanford site, USA, it is necessary to assay the nuclide contents of waste containers by fast methods and non-destructively, employing simple gamma dose rate measurements. The aim of these measurements is to differentiate waste containers in waste category I (according to Hanford Solid Waste Acceptance Criteria) from those that are not suitable for near surface burial. The application of simple dose rate measurements requires that correlations and regions of confidence are known for various waste types. Normally, the wastes contain U, TRU, I 129, Nb 94, C 14, Cs 137, Ba 137, Sr 90, Y 90, Tc 99, Se 79, Nb 93m, Co 60 and Eu 154, where Cs 137 dominates the gamma emitting nuclides. Therefore, correlation factors are calculated for Cs 137. By applying appropriate regions of uncertainty, the use of dose rate measurements allows to characterize waste drums sufficiently, including uncertainties associated with inhomogeneities, composition, and instrument accuracy. Waste drums that do not meet Category 1 limits are analyzed in more detail to determine the waste category.

The results of [FUD 92]/[MCM 94] and [GED 95], respectively, confirm that it is advantageous to characterize decommissioning wastes prior to dismantling (i.e. waste generation) completely in order to be able to perform the subsequent measurements by simple gamma analyses and calculate the activities of difficult to measure nuclides by the previously established "fingerprints". That is common practice in most decommissioning projects. For operating nuclear installations, similar "fingerprints" can be established for various waste streams.

If sufficient knowledge of the process from which the waste has been generated is available, various statistical and even fuzzy evaluations are possible [SMI 94]. Qualitative and quantitative estimates of radionuclides present in a waste stream can be provided can be generated with a computer program that evaluates knowledge of the waste-generating processes as well as limited measurements and that is based upon a fuzzy mathematics model. As an advantage over conventional correlation or other analyses is that the fuzzy model allows to calculate a measure of reliability along with the concentration estimates. Instead of having a deterministic value (single data point), a defined range of probable values can be obtained, from which the potential for a waste package to exceed the applicable limit can be estimated. It must be noted, however, that the origin of the waste package must be known. The advantages of incorporating process knowledge into the radionuclide estimates become less significant if little or nothing is known about the waste generating process.

The results of this work are of little importance for the present study as it concentrates on gas-cooled reactors.



# **Appendix C: Overview of Reports**

# German Reports

Besides the German data base described in [WTI 94a], which is the main basis for German data for this study, additional reports exist in which similar correlation analyses are described. For example, a recent paper is [OPP 93].

### Swedish Reports

# Mätning av Sr 90 och transuranhalter i reaktorvatten och jonbytarmassor (Measurement of Sr 90 and TRU contents in the reactor water and ion exchange resin, SSI P211-83, SKI B18/84) [SSI 84]

This report offers the most comprehensive overview of analyses of alpha and gamma emitting nuclides in the reactor coolant water (Ringhals 1, BWR) with respect to the other reports summarized below. Unfortunately the data are from 1983/84 so that for the present work they are of limited interest.

			Nuclide	contents	in ion e	xchange	resins o	f Swedis	hBWR	[Bq/cm <sup>3</sup>	])		
Re- act.	Waste	Cr 51 - Mn Co 5 54	8 Co 60	Zn:65	Sr.90	-Ag . 110m -	Cs 134	Cs 137	Pu 238	Pu 239	Am 241	Cm 242	Cm 244
B2	Ion ex	1.9e4	5.2e4	1.3e4	7.0e1	3.7e3	0	4.1e3	0	0	0.001	0	0
01	Ion ex	3.2e4	9.3e5	3.0e6	2.8e4	3.0e4	3.9e3	3.2e3	3.74	2.23	2.12	3.77	1.18
R1	Ion ex	1.2e5	2.7e6	1.8e6	8.9e4	9.6e4	2.4e5	2.9e5	1.08	0.484	0.196	2.89	0.108
R1	Ion ex	1.8e5	3.6e6	2.9e6	8.8e4	6.1e4	2.6e5	2.6e5	3.94	2.20	0.50	7.9	0.284
R1	Ion ex	2.0e4	3.1e5	2.6e5	7.5e3	7.9e3	9.5e3	1.2e4	0.42	0.232	30.0	2.75	0.141
R1	Ion ex	1.8e5	5.8e6	4.2e6	1.2e3	2.1e5	1.1e5	1.2e5	6.7	3.32	0.235	2.82	0.24



					Nuclide	contents	in ion é	xchànge	resins o	f Swedis	h BWR	[Bq/cm <sup>3</sup>			
Re-	Waste	Cr 51	(Mn 📎	Co 58	Co 60.	Zn 65	Sr 90.4	Ag	Cs	Cŝ	Pu	Puse	Am	Cm	Cm
act.			54					110m	134	<u>.</u> 137	238	: <u>23</u> 9	241	242	244
R1	Water		0.38	0.17	42	118	2.4e-3	1.2	0.30	0.34	1.8e-5	1.2e-5	9.2e-6	8.9e-5	7.4e-6
R1	Water		0.89	4.3	41	55	5.5e-3	1.24	0.025		1.1e-5	5.4e-6	2.2e-6	2.4e-5	1.8e-6
R1	Water		0.37		17	59	2.4e-3	1.1	1.2	1.5	2.4e-5	1.7e-5	8.2e-6	4.0e-5	5.6e-6
R1	Water		0.14	1.5	18	60	2.5e-3	1.0	0.004	0.063	1.1e-5	5.3e-6	4.1e-6	2.7e-5	2.0e-6
R1	Water		0.58	3.9	26	56	4.5e-3	0.98	0.089		2.9e-5	2.0e-5	8.7e-6	4.5e-5	1.0e-6
R1	Water		1.4	0.67	25	18	1.6e-3	0.19	0.17	0.13	4.0e-5	1.8e-5	5.5e-6	9.3e-5	3.3e-6
R1	Water	300	0.39	4.7	8.2	10	1.4e-4	0.33	0.067	0.042	6.0e-6	2.8e-6	0.9e-6	1.1e-5	0.9e-6
R1	Water		0.23	2.7	3.0	2.1	1.3e-4	0.20	0.029	0.014	3.0e-6	1.8e-6	7.6e-5	1.7e-6	0.3e-6
R1	Water	57	0.45	4.8	3.9	2.5	2.4e-4	0.21	0.077	0.057	3.0e-6	1.8e-6	0.2e-6	1.5e-6	0.2e-6
R1	Water	240	0.51	6.3	8.3	13	0.016	2.2	0.28	0.35	0.4e-6	0.1e-6	1.3e-6	1.2e-5	1.1e-6
R1	Water	83	0.57	6.2	7.4	6.3	3.5e-4	0.52	0.043	0.034	1.3e-6	0.9e-6	1.1e-6	4.4e-5	1.8e-6
R1	Water	18	0.35	5.2	3.9	2.4	2.4e-4	0.31	0.037	8.9e-3	2.5e-6	1.3e-6	4e-8	1.2e-6	9e-8
R1	Water	340	0.66	7.2	8.8	9.5	6.6e-3	1.2	0.075	0.058	6.6e-6	3.9e-6	0.3e-6	3.2e-5	1.2e-6
R1	Water	40	0.37	5.7	4.3	2.5	1.9e-4	0.20	0.019	0.066	3.1e-6	1.8e-6	0.9e-6	6.3e-6	0.1e-6
R1	Water	65	0.33	5.0	3.3	2.0	1.9e-4	0.23	0.010	7.0e-4	1.3e-6	0.9e-6	0.3e-6	8.6e-6	0.5e-6
R1	Water	54	0.26	3.7	2.6	1.5	1.7e-4	0.16	0.019	0.028	1.2e-6	0.8e-6	1.9e-7	5.4e-6	2.2e-7
R1	Water	84	0.40	6.0	4.1	2.4	2.5e-4	0.23	0.028	0.037	2.1e-6	1.3e-6	7e-8	1.9e-6	1e-8
R1	Water	92	0.31	5.2	3.8	2.2	2.3e-4	0.25	0.031	0.021	1.9e-6	1.4e-6	0.0	9.1e-6	3.1e-7
R1	Water	120	0.45	6.6	4.7	2.5	3.3e-4	0.36	0.046	0.017	2.5e-6	1.8e-6	0.0	9.3e-6	1.2e-6
R1	Water	280	0.60	7.2	5.6	3.0	2.9e-4	0.75	0.040	0.12	3.4e-6	1.8c-6	0.2e-6	1.5e-5	0.2e-6
R1	Water	380	0.53	6.2	5.0	3.7	3.7e-4	0.78	0.096	0.071	6.3e-6	4.0e-6	0.8e-6	1.1e-5	0.3e-6
R1	Water	290	0.31	4.1	3.0	1.7	1.3e-4	0.26	0.038	0.059	5.4e-6	2.9e-6	0.4e-6	4.1e-6	0.1e-6
R1	Water	290	0.68	8.9	8.7	4.3	3.9e-4	0.70	0.055	0.054	9.8e-6	4.9e-6	1.7e-6	2.3e-5	1.9e-6
R1	Water	610	0.55	8.2	7.3	6.4	6.9e-3	1.9	0.11	0.10	8.6e-6	5.5e-6	0.6e-6	8.2e-6	0.7e-6
R1	Water	72	0.53	6.2	5.2	2.9	3.1e-4	0.72	0.091	7.4e-3	5.2e-6	1.9e-6	0.2e-6	5.9e-6	0.4e-6
R1	Water	430	1.6	8.0	23	13	5.9e-3	1.8	0.49	0.26	1.4e-5	6.0e-6	0.4e-6	3.4e-5	1.3e-6
R1	Water	340	0.34	5.5	5.3	4.1	5.3e-4	1.5	0.025	0.053	5.6e-6	3.2e-6	0.1e-6	7.6e-6	0.5e-6
R1	Water	54	0.16	2.2	2.2	1.7	1.7e-4	0.29	0.015	0.016	2.0e-6	1.3e-6	0.0	1.8e-6	9e-8
R1	Water	380	0.48	7.0	7.1	5.9	1.1e-3	0.89	0.11	0.21	3.0e-6	1.4e-6	1.4e-6	0.0	-0.0
R1	Water	230	0.53	6.8	7.5	7.1	6.1e-4	0.88	0.29	0.064	1.7e-6	1.0e-6	0.0	3.9e-6	0.4e-6
R1	Water	210	0.46	7.1	6.2	4.0	7.6e-4	0.70	0.057	0.039	3.6e-5	2.4e-5	0.3e-6	1.2e-6	1e-8
R1	Water	130	0.37	4.8	5.4	4.1	1.1e-3	0.49	0.061		2.1e-5	1.6e-5	0.1e-6	4.2e-6	1.0e-6
R1	Water	600	0.57	8.1	7.2	5.4	9.3e-4	0.76	0.087	0.008	2.8e-5	1.7e-5	0.0	6.4e-6	4e-8
R1	Water	340	0.72	8.5	9.0	7.1	1.1e-3	0.86	0.13	0.19	2.8e-6	0.8e-6	2.2e-6	1.3e-5	2.6e-6

# Mätning av transuraner i avfall (Measurement of transuranics in wastes, H.Tovedal, Studsvik NW-85/953, 1986) [TOV 86]

This report deals with measuring methods based on neutron measurements for TRU in conditioned radioactive wastes. Detection limits found in the literature are given. Unfortunately, no measurement data are given so that this report cannot serve for broadening the data base of the present work.

This report can, however, serve as a very good basis for revisiting measurement methods for conditioned waste packages.



Analys av jonbytarmassor från Oskarshamn III med avseende på transuraner och gammastrålare (Analysis of ion exchange resins from Oskarshamn III with regard to TRU and gamma emitting nuclides, SSI P2-87, Mats Skålberg) [SKA 87]

This report describes the analysis of ion exchange resins with respect to TRU and  $\gamma$ -emitting nuclides. Gamma spectrometry and alpha measurements have been used and the measurement methods are described in detail.

The results of the measurements are reproduced in the table below. An important fact is that apart from the alpha emitting nuclides also gamma emitting nuclides have been analysed so that a correlation analysis with Cs 137 or Co 60 is possible.

()ynges	Zr 95	Ru 103	Ru 106	Ag 110	Sb 124	Sb 125	Cs 134	Cs 137	Ce 144	Co 58	Co 60	Zn 65	Mn 54	Fe 59	Pù 239*	Pu 238	Am 241	Cm 244	Cm 242
1				2.0E+ 03			2.94E +04	7.55E +04		8.24E +5	2.39E +05	1.76Ē +04	1.49E +05						
3.20								4.08E +00	1.20E +01		6.15E +01		4.51E +01						
5	4.03E +05	5.53E +05	1.56E +05	9.00E +03	1.36E +05	1.70E +4		3.00E +03	2.93E +5	4.55E +6	3.25E +6	3.80E +05	1.09E +06	8.90E +05					
<b>6</b>	1.00E +04	3.00E +03	9.00E +03	4.00E +02		4.00E +02	5.00E +03	1.30E +04	6.00E +03	1.70E +04	2.60E +04	2.00E +03	1.10E +04	7.00E +02					
12.00	6.00E +03					4.00E +03	5.00E +03	1.20E +04		1.00E +04	1.40E +04	1.00E +03	5.00E +03						
8	3.00E +03		1.00E +02			1.00E +02			1.00E +03	1.00E +04	1.80E +04	2.00E +03	6.00E +03						
9									1.00E +03	4.00E +03	3.00E +03	3.00E +02	1.00E +03						
A															7.4E+ 00	1.38E +00	3.30E -02	5.5E- 03	5.8E- 01
Sa	mples	i, 3 and	A: Bq	/g; Sar	nples 5	-9: Bq													-
*)	Pu 239	= Pu2	39+Pu2	240															ļ

# SFR - Redovisning av TRU och Sr 90 1988-91 (SFR - report about transuranics and Sr 90 1988-91, SKB, 1993-01-12, Marie Johansson) [JOH 93]

In this report it is stated that a working group with delegates from the NPPs and SKB has worked out principles for the assessment of TRU and Sr 90 in reactor wastes. The methods used are based in the first place on the analysis of the reactor water and the water of the storage pools that are the dominant sources of TRUs while the TRU contents of the condensate demineralizer system and the system drainage contribute only a fraction of this.

Detailed lists of measurements in the NPPs Oskarshamn, Ringhals, Forsmark and Barsebäck are given for the years 1988 until 1991 (CLAB is included also). In the following list the annual totals for the NPPs are reproduced.



		an a	Nuclide	specific	measure	ments (S	r 90 and	TRU) o	f the was	tes of Sy	vedish NPPs [B	9] <u>``</u>
NPP	year	Sr.90,	U 235	U 236	Np 237	U 238	Pu 238	Рц 239	Am 241	Cm 242	Am Cm 243 243	Cm 244
OKG	1988	3.58E +11		1.30E +06	1.60E +06	4.30E +06	1.14E +08	3.99E +08		3.60E +08	2.50E +06	1.92E +07
	1989	2.31E +10	3.20E +04	3.00E +04	7.60E +04	3.30E +04	7.96E +07	1.02E +08		4.95E +08		2.50E +07
	1990	1.18E +10		2.00E +04			7.17E +07	2.98E +07		5.72E +08		3.38E +07
	1991	1.14E +10					9.69E +07	3.44E +07		4.24E +08	4.60E +05	5.72E +08
BVK	1988	4.63E +09						3.59E +04				
FVK	1988	2.37E +11	2.82E +05		4.64E +06	9.98E +05	9.57E +05	8.39E +07		5.78E +08		2.08E +07
	1989	2.18E +11	4.15E +05		1.52E +06	1.54E +05	3.08E +08	2.89E +08	2.39E +08	7.04E +08		4.72E +07
	1990	6.93E +10					1.22E +08	4.79E +07		4.01E +08		9.14E +07
	1991	1.13E +12	2.53E +06		3.00E +07	3.16E +06	8.30E +08	4.38E +08	1.50E +07	2.56E +09		2.88E +08
RVK	1988	3.42E +10					2.30E +09	1.16E +09	4.62E +08			9.10E +08
	1989	2.04E +10					3.34E +08	2.17E +08	1.56E +08			4.52E +08
	1990	3.86E +10					1.08E +09	3.10E +08	8.15E +08			1.05E +09
	1991	1.33E +10					2.25E +08	5.48E +07	1.23E +08			3.20E +08

### BKM-G1- Sammanställning av utförda bränsleskrapsundersökningar (BKM-G1 - Compilation and conduction of fuel investigations, 1990, ABB Atom) [ABB 90]

This report contains detailed information about the chemical composition of materials related to the fuel elements and the chemical behaviour of various elements (Co, Cr, Cu, Fe, Mn, Ni, Zn). It does not contain however any nuclide vectors. So in the context of this study it will be used only as background information in a later stage.

# An Assessment of Pu 238 and Pu 239+240 in the primary cooling water of a PWR (SFR, Progress Report Risø-R-619, Feb. 1992, Q. Chen, S.P.Nielsen, S.Duniec) [CHE 92]

In this report the Pu contents of the primary cooling water is analyzed first by radiochemical preparation and then by alpha-spectrometry. The samples of the cooling water have been taken from Ringhals 2 at points before and after the ion-exchange system. The concentrations of Pu 238 and Pu 239+240 have then been determined. As the main aim of the work presented in this report was the test of the efficiency of the ion exchange system for removal of Pu from the reactor water,



the results presented can only be of limited value for the present study. Only the measured concentrations for Pu 238 and Pu 239+240 are given, no additional measurements of Cs 137 or any other nuclide are presented so that these data cannot form the basis for a correlation analysis. The data can only serve as a basis for a correlation between Pu 238 and Pu 239+240.

Samples	s before ion exc	change system	[mBq/l]	Samples after ion exchange system [mBq/l]					
Pu 238	Pu 239+240	Pu 238	Pú 239+240	Pu 238	Pu 239+240	Pu 238	Pu 239+240		
128.0	34.0	7.2	3.7	7.2	3.3	1.2	0.4		
12.0	4.0	9.0	3.6	1.7	1.2	10.6	4.0		
21.0	5.6	4.8	1.8	4.2	1.3	8.4	4.2		
45.0	22.0	8.1	3.9	8.2	4.9	3.4	1.0		
20.0	7.1	2.5	1.1	75.5	24.0	6.1	2.1		
401.0	300.0	2.9	1.1	3.2	2.0	6.3	2.4		
2.6	0.7	8.4	3.8	14.6	6.1	2.7	1.0		
1.3	0.9	8.4	3.1	11.7	7.5	1.0	0.3		
1.0	0.7	3.9	1.1	277.9	43.6	1.1	0.3		
7.4	15.0	9.8	4.3	1.7	1.3	0.8	0.1		
3.4	3.2	11.9	5.0	2.7	1.2	2.7	1.0		
2.6	0.6	6.8	7.8	5.8	2.6	2.5	1.4		
27.0	12.2			3.3	0.9	2.0	1.0		
17.6	5.3			1.7	0.6	1.6	0.7		

# Strontium 90 och transuraner i reaktoravfall (Sr 90 and TRU in reactor wastes, Vattenfall Ringhalsverket)

The series of reports dealing with Sr 90 and TRU contents in wastes comprises the following reports:

- Strontium 90 och transuraner i reaktoravfall 1988 och 1989 (Sr 90 and TRU in reactor wastes 1988 and 1989, Vattenfall Ringhalsverket, Rapport 276/90, 1990) [VAT 90]
- Transuraner och Strontium 90 i reaktoravfall 1990 (TRU and Sr 90 in reactor wastes 1990, Vattenfall Ringhalsverket, Rapport 364/91, 1991) [VAT 91]
- Transuraner och Strontium 90 i reaktoravfall 1991 (TRU and Sr 90 in reactor wastes 1991, Vattenfall Ringhalsverket, Rapport 0599/92, 1992) [VAT 92]

In each report analyses of Sr 90, Pu 238, Pu 239/240, Am 241 and Cm 244 is given for various sampling locations. Unfortunately, no measurements of Co 60 or Cs 137 are included so that a further examination of these reports is postponed.

Ringhalsverket årsrapport för 1992 transuraner och Sr-90 i reaktoravfall (Ringhals plant annual report for 1992: TRU and Sr-90 in reactor waste. Vattenfall Ringhalsverket, Rapport 0794/93, 1993) [VAT 93]

This report shows the results of sampling (Sr 90 and TRU) during reactor, condenser and fuel element basin cleaning. The measurements are carried out four times a year comprising the reactor water and the storage pool water. The measured nuclides comprise Sr 90, Pu 238, Pu 239+240, Am 241 and Cm 244 for Ringhals 1-4. The data are summarized in the table.



an a the constant of the second s Second second	$\overset{\text{def}}{\underset{k \in \mathcal{K}}{\overset{\text{def}}{\underset{k \in \mathcal{K}}{\overset{\text{def}}}{\underset{k \in \mathcal{K}}{\overset{\text{def}}{\underset{k \in \mathcal{K}}{\overset{\text{def}}{\underset{k \in \mathcal{K}}{\overset{\text{def}}{\underset{k \in \mathcal{K}}{\overset{\text{def}}}{\underset{k \in \mathcal{K}}{\overset{\text{def}}}{\underset{k \in \mathcal{K}}{\overset{\text{def}}}{\underset{k \in \mathcal{K}}{\overset{\text{def}}}{\underset{k \in \mathcal{K}}}{\underset{k \in \mathcal{K}}{\underset{k \in \mathcal{K}}{\atop\atopk \\{k \in \mathcal{K}}}{\underset{k \in \mathcal{K}}}{\underset{k \in \mathcal{K}}}}{\underset{k \in \mathcal{K}}{\underset{k \in \mathcal{K}}}{\underset{k \in \mathcal{K}}}{\underset{k \in \mathcal{K}}}}}}}}}}}}}}}}}}}}}}}}}}}}}}}}}}}}$	Activity [Bq] per	nuclide in the io	n exchange resin	S
Ringhals	Sr 90 [Bq]	Pu 238	Pu 239+240	Am 241	Cm 244
1	4.64E+09	1.08E+07	1.33E+07	8.21E+06	1.78E+07
2	6.03E+09	2.32E+07	8.95E+07	2.81E+07	9.85E+07
3	1.69E+09	3.65E+06	4.66E+06	5.27E+06	5.73E+06
4	1.06E+09	2.20E+06	2.78E+06	1.95E+06	3.47E+06
Sum	1.34E+10	3.99E+07	1.10E+08	4.35E+07	1.25E+08

In addition, correlation factors have been calculated for the above mentioned nuclides relative to Co 60 (no reason for a correlation with Co 60 instead of Cs 137 is given) which are reproduced below:

	Correlat	ion factors relat	ive to Co 60 in	the ion exchange	ge resins
Reactor	Sr 90 [Bq]	Pu 238	Pu 239+240	Am 241	Cm 244
R1, reactor cleaning	1.18E-03	1.27E-06	2.07E-06	1.59E-06	2.97E-06
R1, storage pool	6.00E-05	1.60E-06	1.48E-06	6.11E-07	1.78E-06
R2, reactor cleaning	1.49E-02	1.79E-05	4.59E-05	7.60E-06	2.16E-06
R2, storage pool	1.14E-03	1.18E-05	4.99E-05	1.70E-05	6.04E-06
R3, reactor cleaning	1.28E-02	5.90E-06	1.05E-05	3.05E-05	3.00E-06
R3, storage pool	4.13E-04	3.61E-06	4.23E-06	2.45E-06	3.07E-06
R4, reactor cleaning	3.94E-03	5.77E-06	7.11E-06	4.45E-06	1.12E-06
R4, storage pool	3.29E-04	1.77E-06	2.33E-06	1.87E-06	1.86E-06
mean	4.34E-03	6.21E-06	1.54E-05	6.25E-06	1.60E-06

Nuklidspecifika alfa-analyser (Nuclide specific alpha analyses, OKG, Rapport 8-804/87, 1987)

In this report some data for alpha emitting nuclides are given. Direct measurements of Cs 137 and Co 60 are missing, so that these data cannot serve directly for correlation analyses. The data are reproduced in the following table.



Cher Hay State	hardan di kada	New Sold	Contra go A	Upha analy	ses from c	ooling wat	er [Bq/cm <sup>3</sup>	<b>]</b>	Sister A.
Reactor	Waste	Pu 239/	Am 243	Pu 238	Cm 244	Cm 242	Alpha	Alpha	Co 60
al alarasi	a start and a start	240	de des corto	Am 241	a de la calificia	an The Alam Sant	?tot:	tot (2).	S Martha St.
B1		4.6E-04	3.2E-04	1.8E-03	1.3E-03	2.2E-03	6.4E-03	3.7E-03	3.4E+02
B2								3.1E-03	7.6E+02
F1		4.7E-04	2.2E-04	2.1E-03	1.6E-03	3.4E-03	7.8E-03	2.7E-03	1.1E+03
F2								3.6E-03	9.8E+02
01	Water	2.8E-03		1.1E-02	1.2E-02	3.6E-02	6.2E-02	2.1E-02	5.6E+02
02								7.9E-03	2.1E+02
R2			7.6E-04	1.1E-03	6.8E-04	7.2E-04	3.3E-03	1.3E-02	1.7E+03
R3								6.1E-03	1.5E+03
R4								5.1E-03	2.9E+02
KWO	í l	9.6E-04		2.3E-03	8.2E-04	3.9E-03	8.8E-03		

### CLAB - Uppskattning av transuraninnehåll i avfall (CLAB - Assessment of the TRU contents in the wastes)

The series of reports dealing with TRU contents in wastes based on activity analyses of the storage pool water or the water from the internal cooling system at CLAB comprises the following reports:

- CLAB Uppskattning av transuraninnehåll i avfall från 1988 (CLAB Assessment of the TRU contents in the wastes of 1988, OKG, Report 8-473/89, 1989)
- CLAB Uppskattning av transuraninnehåll i avfall från 1989 (CLAB Assessment of the TRU contents in the wastes of 1989, OKG, Report 8-103/90, 1990)
- CLAB Uppskattning av transuraninnehåll i avfall från 1990 (CLAB Assessment of the TRU contents in the wastes of 1990, OKG, Report 91-03206, 1991)

A very important statement that can be found in all three reports is that the TRU contents correlates with the Co 60 contents in the waste packages.

The following table shows the data as extracted from the reports.



ingen solgen geste Standarder solgen	ر به دمین رو معرفی مراجع می	an fill a star	na star na star (n Seneral an star (na st	Nuclidec	ontents of v	vàstes [Bq]		en e
Waste	year 🔅 🔅	Pü 238/	Pu 239/	Cm 242	-Cm 243	Cm 244	Am 243	Co 60
and a company and the second	a an	Am 241	240		t Staling States	<u>na plata a</u>	and the second	
Intern.	1988	4.3E+04	1.2E+04	2.5E+04	3.6E+04	3.1E+03	3.0E+02	1.45E+10
cool. syst.								
		1.7E+04	4.9E+03	9.7E+03	1.4E+04	1.2E+03	1.0E+02	5.8E+09
	1989	1.4E+04	7.8E+03	7.9E+04	7.2E+03	1.5E+04	2.5E+03	3.8E+10
		7.6E+03	4.4E+03	4.4E+04	4.1E+03	8.5E+03	1.4E+03	2.1E+10
	1990	8.1E+04	2.3E+04		2.3E+04	2.1E+04	1.9E+03	1.7E+10
		1.4E+05	4.0E+04		4.0E+04	3.5E+04	3.2E+03	2.9E+10
Filter/ion	1988	6.0E+03	1.7E+03	3.5E+03	5.0E+03	4.0E+02	4.0E+01	2.0E+09
ex. resin	1989	2.1E+03	1.2E+03	1.2E+04	1.1E+03	2.4E+03	4.0E+02	6.0E+09
	1990	2.2E+04	6.3E+03		6.3E+03	5.6E+03	5.0E+02	4.6E+09
Filter	1988	1.0E+05	4.0E+04	8.0E+04	1.0E+05	1.0E+04	1.0E+04	4.7E+10
		6.0E+05	2.0E+04	4.0E+05	5.0E+05	5.0E+04	0	2.1E+11
	1989	4.0E+04					3.0E+04	1.8E+11
		5.0E+04					4.0E+04	2.3E+11
		7.0E+03					6.0E+03	3.4E+10
		4.0E+04					2.0E+04	9.3E+10
	1990	1.3E+07	3.4E+06			3.8E+06	2.5E+05	5.5E+11
		1.3E+07	6.1E+05			8.0E+04		5.0E+11
		1.2E+06				7.0E+04		1.0E+11
		1.3E+06	1.0E+04			4.0E+04		1.3E+11
Filter	1988	3.1E+06	8.0E+05	6.0E+04	2.4E+06	4.0E+06	5.0E+05	1.4E+13
		2.6E+06	6.0E+05	5.0E+04	1.9E+06	3.3E+06	4.0E+05	9.2E+12
	1989	9.2E+06	1.4E+06	2.0E+04	9.7E+05	3.4E+06	3.6E+06	9.1E+12
		9.2E+06	1.4E+06	2.0E+04	9.7E+05	3.4E+06	3.6E+06	8.0E+12
	1990	4.9E+07	1.4E+07			6.0E+06	6.2E+05	1.8E+13
		9.4E+06	2.8E+06			1.1E+06	1.2E+05	4.0E+12

### Svårmätbara betastrålande radionuklider - Ni 59, Ni 63 och Tc 99 (Difficult to measure radio nuclides) SKB PM 94/19, A4522, 1994 [ROO 94]

In this report, the measurement results of 48 reactor water samples are reported. Measurements have been performed for Ni 59, Ni 63, Te 99, Co 60 and Cs 137. Procedures for the chemical separation of Ni and Tc are described.

Reactor	Date 20	Có 60 [Bg/kg]	Ni 59 [Bq/kg]	Ni 63 [Bq/kg]	Tc 99 [Bq/kg]	Cs 134 [Bq/kg]	Cs 137 [Bq/kg]
01	1989	5.32E+03		9.00E+01	1.10E-01		7.70E+03
01	1990	6.85E+03		9.40E+01	7.50E-02		2.25E+01
01	1990	2.55E+02		5.20E+01	1.41E+00		8.46E+03
O2	1989	3.50E+03		2.80E+02	1.57E+00		4.40E+04
02	1989	4.16E+03		3.50E+02	1.42E+00		3.27E+04
02	1989	3.69E+03		3.20E+02	1.58E+00		5.21E+04
O2	1989	4.23E+03		3.80E+02	2.30E-01		1.04E+03
O2	1990	5.05E+03		3.80E+02	2.20E-01		1.10E+03
O2	1990	5.58E+03		2.90E+02	4.50E-01		7.20E+03
02	1990	5.32E+03		4.20E+02	4.28E+00		4.71E+05
O2	1990	5.09E+03		1.60E+02	2.60E-01		3.64E+03



Reactor	inter Ski	Co 60 [Bq/kg]	Ni 59 [Bq/kg]	Ni 63 [Bq/kg]	Tc 99 [Bq/kg]	Cs 134 [Bq/kg]	Cs 137 [Bq/kg]
02	1989	2.80E+02		8.60E+01	4.70E-01		2.35E+03
02	1989	5.00E+01		2.20E+01	2.20E-01		4.40E+02
02	1990	5.10E+01		1.70E+01	3.50E-01		7.00E+02
02	1990	4.30E+01		1.50E+01			
02	1990	2.95E+02		1.07E+02	4.80E-01		2.88E+03
02	1990	9.00E+01		4.30E+01	1.00E+00		9.00E+03
03	1989	6.94E+03		9.90E+01			
03	1990	5.90E+03		8.60E+01			
B1	1989	7.60E+03		1.85E+02	1.34E+01	· · · · · · · · · · · · · · · · · · ·	1.61E+07
B1	1989	1.95E+03		1.49E+02	1.00E-01		2.00E+01
B1	1989	5.35E+02		5.50E+01	1.00E-01		1.50E+02
B1	1990	3.85E+02		5.40E+01	1.00E-01		1.20E+02
B1	1990	2.40E+02		2.80E+01			
B1	1990	2.71E+03		3.10E+01	1.00E-01		2.00E+03
B1	1990	2.20E+02		3.20E+01			
R1	1989	3.18E+03		3.50E+01	4.90E+00		
R1	1989	2.70E+03		1.06E+02	9.00E-01		1.80E+02
R2	1989	1.25E+02		3.70E+01			
R2	1989	1.54E+03		8.80E+02	1.10E+00		7.70E+02
R2	1989	9.84E+04	3.80E+01	5.43E+04	2.20E+00		6.60E+03
R2	1989	1.68E+03		7.60E+02	3.80E+00		1.90E+04
R2	1989	3.43E+03		1.57E+03	6.50E-01		5.20E+02
R2	1989	1.02E+04		4.85E+03	2.00E+00		9.80E+03
R2	1989	1.73E+04	6.30E+00	8.26E+03	1.70E+00		7.99E+03
R2	1989	2.51E+04		1.28E+04	4.10E+00		2.75E+04
R3	1989	4.05E+03	1.30E+01	1.60E+04	8.00E-01		4.80E+02
R3	1989	8.45E+03	1.00E+01	8.24E+03	9.00E-01		9.00E+03
R4	1989	1.04E+03	4.50E+00	5.69E+03	1.70E+00		1.70E+02
R4	1989	3.20E+03	5.20E+00	7.42E+03	5.00E-01		7.50E+02
CL	1989	6.84E+03		1.54E+03	6.00E-01		1.02E+04
CL	1989	7.01E+03		2.17E+03	1.60E-01		8.00E+02
F1	1990	8.60E+03		1.80E+02	1.94E+00	490	9.70E+02
F1	1990	8.80E+01		4.80E+01	2.00E-01	265	6.00E+02
F1	1990	8.59E+04		2.23E+03	1.23E+01	4230	7.58E+03
F1	1990	3.32E+04		2.06E+03	3.90E+00		
F1	1990	9.20E+03		1.17E+03	1.60E+00		3.60E+02
F1	1990	9.00E+02		1.40E+01	5.00E-02		

# **USA Reports**

The US data are mainly contained in two reports. The first and most comprehensive report is

• NUREG/CR-4101: Assay of long-lived radionuclides in low-level wastes from nuclear power plants, 1985

which contains data from a great variety of NPPs until approximately the year 1985.

More recent data (but less comprehensive) can be found in the report



• DOE/RW-0184-R1: Characteristics of potential repository wastes, U.S.DOE, 1992

These two reports are included in the analysis.

### Japanese Reports

The Japanese data are taken from the study "Methods of Determining the Activity Concentration in Radioactive Waste" (Federation of electric power companies, Tokyo, 1989). Among other topics, this report summarizes various correlation analyses performed for Japanese NPPs. These analyses were part of research work for the confirmation of activity concentrations of low-level radioactive waste bound for shallow land disposal.

The report distinguishes between three methods for the data analysis: 1.) the actual measurement for each waste package which is a very complex method especially for the difficult to measure nuclides; 2.) the scaling factor method which mainly corresponds to a normal correlation analysis described above and 3.) the theoretical analysis method which derives the estimated contents of the corrosion and fission products from the individual formation pathways and the release into the cooling water or other contaminants. In the context of this report, method 2 is of the greatest importance as it is based on actual measurements and can be compared directly to the measurements and correlations performed with the data of the other countries.

The key nuclides that have been chosen are Co 60 and Cs 137. The following correlations have been performed:

- Ni 59, Ni 63, Nb 94 with Co 60
- Ni 59 with Ni 63
- Sr 90, I 129, total alpha with Cs 137
- no correlation found for C 14 and Tc 99

It should be noted that the correlation analyses are performed on the actual measurement data and not on the logarithm of these data, i.e. the resulting correlation equation is of the form

 $A_{\text{Nuclide difficult to measure}} = a \cdot A_{\text{Key nuclide}} + b$ 

# **Appendix D: Compilation of Information for each Nuclide**

In this appendix nuclide specific information is listed. Table 1 [WTI 94a] gives an overview of the formation pathways of various nuclides relevant for wastes from nuclear power plants. It is provided as a quick reference for the half-life and for the possible origin (activated corrosion product, activated concrete, contamination, fuel etc.) of each nuclide. More details can be found in the subsequent nuclide specific chapters.

Nüclide	Half-life	Fission product	From thermal neu-	From fast neutrons	Others	From UO <sub>2</sub>
	ි [a]ළැදු	or natural decay	trons		8855.43	
H 3	$123.10^{1}$	and the second se	Li6 H2	Contractory ( Contractory Cont	Li6	LE SP
C 14	$5.74 \cdot 10^3$		C 13	O 17. N 14		LE
Na 22	$2.60.10^{\circ}$		0.10	Na 23	Mg 25	
Cl 36	3.00·10 <sup>5</sup>		Cl 35	K 39. CI 37		(C)
Ca 41	$1.30.10^{5}$		Ca 40	Ca 42		
Mn 54	$3.12 \cdot 10^2$		~~	Mn 55. Fe 54	V 51	(V)
Fe 55	2.70·10 <sup>0</sup>		Fe 54	Ni 59	Fe 54	
Ni 59	7.50.10⁴		Ni 58	Ni 60		
Co 60	5.27.10 <sup>0</sup>		Co 59	Ni 60. Cu 63		
Ni 63	$1.00.10^{2}$		Ni 62	Zn 66. Cu 63		
Se 79	6.50·10 <sup>4</sup>		Se 78	Kr 82. Br 79		FP. (Br)
Kr 85	$1.07 \cdot 10^{1}$	U 235	Kr 84	Rb 85		FP
Sr 90	2.85·10 <sup>1</sup>	U 235				FP
Mo 93	3.50·10 <sup>3</sup>		Mo 82	Ru 96, Mo 94		(Ru), (Mo)
Zr 93	1.50·10 <sup>6</sup>		Zr 92	Zr 94, Nb 93		FP
Nb 94	2.03.10⁴	U 235	Nb 93			(Nb)
Nb 95	9.63·10 <sup>-2</sup>	U 235	Zr 94	Mo 93		FP, (Mo)
Zr 95	1.76·10 <sup>-1</sup>	U 235	Zr 94	Mo 98		FP, (Mo)
Тс 99	2.14·10⁵	U 235	Mo 98			FP
Pd 107	6.50·10 <sup>6</sup>		Pd 108	Cd 110, Ag 107		FP
Ag 108	1.27·10 <sup>2</sup>		Ag 107		Ag 107	?
Cd 113	1.46·10 <sup>1</sup>	U 235	Cd 112	Cd 113		FP
I 125	1.60·10 <sup>-1</sup>		Xe 124	Xe 126	Sb 123	?
Sn 126	1.00·10 <sup>5</sup>	U 235				FP
I 129	$1.57 \cdot 10^{7}$	U 235	Te 128	Xe 129		FP
Ba 133	$1.07 \cdot 10^{1}$		Ba 132	Ce 136, Ba 134		?
Cs 134	$2.06 \cdot 10^{\circ}$	U 235	Cs 133	Ba 134		FP
Cs 135	2.00.12	U 235				FP
Cs 137	3.01·10 <sup>1</sup>	U 235	Xe 136	Ba 136		FP
Sm 151	9.30·10 <sup>1</sup>	U 235	Sm 150	Eu 151		FP
Eu 152	1.27.10		Eu 151	Gd 152, Eu 153		FP
Eu 154	8.50·10°	U 235	Eu 153	Gd 154		FP
Eu 155	4.96∙10°	U 235	Sm 154	Gd 155		FP

 Table 1
 Possible sources of radionuclides in light water reactors



Nuclide'	Half-life	Fission product	From thermal neu-	From fast neutrons	Others	From UO2			
	, [a] ,	or natural decay.	trons						
РЬ 210	$2.04 \cdot 10^{1}$	U(nat) 238				AC			
Ra 228	5.75·10⁰	Th(nat) 232				AC			
Ac 227	2.18·10 <sup>1</sup>	U(nat) 235			ł	AC			
Ac 228	7.00 10-4	Th(nat) 232				AC			
Pu 241	$1.52 \cdot 10^{1}$		Pu 240			AC			
Ra 224	$1.00 \cdot 10^{-2}$	Th(nat) 232				AC			
Ra 226	$1.60 \cdot 10^{3}$	U(nat) 238				AC			
Th 228	1.91·10 <sup>0</sup>	Th(nat) 232				AC			
Th 230	7.70.10⁴	U(nat) 234				AC			
Th 232	$1.41 \cdot 10^{10}$	Th(nat) 232							
Pa 231	3.28.10⁴	U(nat) 235				AC			
U 232	$7.20.10^{1}$			U 233	Th 232	AC			
U 234	2.48·10 <sup>5</sup>	U(nat) 238	U 233	U 235		AC			
U 235	$7.04 \cdot 10^{8}$	U(nat) 235			[	AC			
U 236	2.34.10		U 235			AC			
U 238	4.70·10 <sup>9</sup>	U(nat) 238			1	AC			
Np 237	2.14·10°			U 238		AC			
Pu 239	2.41.10⁴		U 238			AC			
Pu 240	$6.54 \cdot 10^{3}$		Pu 239			AC			
Pu 242	3.87·10 <sup>°</sup>					AC			
Am 241	$4.33 \cdot 10^2$		Pu 240			AC			
Am 242	$1.52 \cdot 10^2$		Am 241			AC			
Am 243	$7.38 \cdot 10^3$		Am 241			AC			
Cm 245	8.50·10 <sup>3</sup>		Pu 239			AC			
Cm 246	4.82·10 <sup>3</sup>					AC			
Cm 248	Cm 248   3.61·10 <sup>3</sup>   AC								
AC = actinides, $FP = fission$ product, $LE = light$ elements, () = impurities in the fuel									

In the subsequent nuclide specific chapters, the following relevant information are listed for each nuclide:

- 1. Properties and formation of the nuclide
  - Half-life,
  - Decay mode(s),
  - Most relevant decay data,
  - Formation pathway and sources in nuclear power plants.
- 2. Measurement

- Measurement techniques for the nuclide as described in the literature.
- 3. Data and correlation
  - Description of available data,
  - Reasons for selection of key nuclide,



• Correlations (if data are available).

### 4. Discussion

• Discussion of the results of the correlation analysis as appropriate.

The decay data at the beginning of each nuclide chapter only show the most important decay modes and energies for the nuclide (if there are more than one). These information, together with the short description of possibilities for measuring the nuclide after separation, allow the reader for example to choose suitable detectors and to asses difficulties of the measurement. The four columns listed for each nuclide are reproduced from [ICR 83] and have the following meaning:

1.	Radiation:	Type of radiation being considered
2.	y(i) [Bq⋅s] <sup>-1</sup> :	Corresponding mean number of events per nuclear transformation. The unit, $[Bq \cdot s]^{-1}$ , is equivalent to transformation <sup>-1</sup> .
3.	E(i) [MeV]:	Corresponding transition energy in million electron volt [MeV].
4.	y(i) E(i):	Product of values in columns 2 and 3: y(i) E(i). Average energy emitted per

transformation for the associated radiation type specified in column 1.

The diagrams with the correlation analyses are shown in a 1:1 axis ratio at the end of each nuclide chapter. In these diagrams the details about the statistical analysis are plotted at the bottom right corner. Together with the equations of appendix A these information allow the assessment of the quality of the correlation and the calculation of the prediction interval (region of confidence). Relevant results of these statistical data are discussed in the text. Where correlation analyses have been performed using data from several countries, only the correlation plot with all data is shown (e.g. for Ni 63 and PWR data from Sweden, Germany, USA and Japan have been plotted in one diagram). In an additional table correlation data are given for each country in order to assess the correlation quality separately. In order to assess the quality of a correlation easily from the various parameters, the meaning and typical value ranges are repeated from Annexe A in the following table. The value ranges for correlation with good quality are meant only for a quick orientation and should not be regarded as strict limits. Often this depends on the additional factors like number of data points etc.

Appendix D



Para- meter	Meaning	value range for correlation with good quality	unacceptable value range-
a	slope of regression line - expo- nent in $A_1 = b \cdot (A_2)^a$	(not applicable)	(not applicable)
b	intersection of regression line with y axis - pre-factor $b' = 10^{b}$	(not applicable)	(not applicable)
	correlation coefficient	≈ 0.8 - 1.0	< 0.5
H	quantile of F distribution, $F_{0.025;1;n}$ , must be compared with critical F value $F_{crit}$ , usually $F_{crit} \leq 1$	depending on number of data points ca. F ≥ 50 200	depending on number of data points ca. $F \le 1 10$
Sxy	residual variance, calculated from y values of data pairs $(y_i)$ and corresponding value $3$ of the regression line	depending on number of data points ≈ 0.3 ≈ 0.7	(not applicable)
Q	sum of squares of deviations of x values from mean $(\bar{x})$	depending on number of data points	(not applicable)
n K	number of data points	data set needs at least sev- eral data points	less than $\approx 5$

In many tables, standard computer scientific notation is used. For example, 1.23e1 or 1.23E1 means the same as  $1.23 \cdot 10^{+1}$ .



# 1. Properties and Formation

Τ <sub>Ά</sub>		12.3 a	
Decay mode:		$\beta^{-}: {}_{1}^{3}H \xrightarrow{\beta^{-}} {}_{2}^{3}He$	
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	y(i) • E(i)
β <sup>-</sup>	1.0	5.683E-03	5.68E-03

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

H 3 is an activation product (thermal neutrons) from Li 6 and Li 7 (the part of Li 7 is negligible because of the small cross section). A formation from deuterium with thermal neutrons is also possible but is only relevant for reactors moderated with heavy water. H 3 is also a fission product from  $UO_2$ . Sources for H 3 are irradiation of B and Li in the absorber material on control elements, the fuel elements and B and Li in the coolant

## 2. Measurement

An analytical technique for the measurement of tritium is described in [NUR 85]:

Tritium is separated and purified from activation and fission product activities by distillation. An alkaline permanganate mother liquor is used to provide an oxidizing environment. The halogens and other elements are rendered non-volatile and remain in the mother liquor. The purified tritium is collected as the distillate. The tritium is prepared for counting in the liquid scintillation counter. A liquid scintillation counter is required for counting.

## 3. Data and Correlation

The activity of H 3 in waste cannot be determined by correlation with key nuclides as H 3 - unlike fission and corrosion products - is not enriched in ion exchange resins or in concentrates. Instead, H 3 can be found in wastes if they contain moisture that is contaminated with water from the primary coolant loop. Therefore, the H 3 concentration can only be estimated from the percentage of primary coolant in the wastes. The analysis has to differentiate between various waste streams.

Data are only available from Germany. The plots show tentative correlations of H 3 with Cs 137 and Co 60. It can be seen that there is indeed no correlation between these nuclides. No data for

the moisture contents of the waste packages are available. Therefore, the correlation between the activity of H 3 and the moisture contents cannot be investigated.

The poor qualities of the correlations can also be seen from the low values of r and F that show that the correlations are statistical not relevant.

### 4. Discussion

It can be assumed that tritium plays no role in the contamination on steel and concrete. According to [SMI 85], there should be no deposition of H 3 on metallic surfaces. Tritium will diffuse further into concrete than other nuclides, but on the other hand the absorbed fraction of H 3 on concrete should be rapidly desorbed. Consequently, wastes arising from contaminated metal or concrete should contain only negligible amounts of tritium.

For other kinds of waste, the tritium contents has to be determined e.g. from the moisture contents. The approach via correlation analyses with Co 60 and Cs 137 does not seem possible.

Correlation between H 3 and Co 60





Correlation between H 3 and Cs 137





# 1. Properties and Formation

Tiz		5730 a	
Decay mode:		$\beta^{-}: {}^{I_4}_{6}C \xrightarrow{\beta^{-}} {}^{I_4}_{7}N$	
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	<b>y(i) · E(i)</b>
β.	1	4.945E-02	4.95E-02

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

C 14 is produced through fast neutron reactions on N 14 and O 17 in the coolant and components and by activation of oxygen in  $UO_2$ . It can also be produced by activation of C 13 (thermal neutrons) in components and concrete. Most of the activity is released in gaseous form, but some of it will be present in the waste in insoluble form.

## 2. Measurement

An analytical technique for the measurement of C 14 is described in [NUR 85] as follows:

Oxidizing distillation technique is employed for liquid or solid samples, which converts more than 97 % of all carbon species to carbon dioxide. The  $CO_2$  is absorbed in a liquid scintillation cocktail and analyzed for C 14 content. A liquid scintillation counter is required for counting.

Additional information can be found in [NCR 85]:

The first step is the transfer of carbon to  $CO_2$  as described above. A common technique for the following introduction of  $CO_2$  into a scintillation liquid is the absorption of  $CO_2$  by a base, e.g. phenethylamine or ethanolamine, that is subsequently dissolved in the proper mixture. This scintillation liquid can then be introduced into the counter.

## 3. Data and Correlation

A correlation with Co 60 can be expected. In [NUR 85] it is suggested that C 14 may be present as carbonate. Therefore, C 14 can be present in wastes from PWRs where the water chemistry can be slightly basic and insoluble carbonates can be formed. The US data however show that also in BWRs C 14 is present.

Data are available from Germany (mostly PWR), USA (both PWR and BWR) and Japan (only BWR). The following plot shows the correlation analysis for C 14 and Co 60.

The correlations are of the form:

 $A_{C 14} = 0.3 \cdot (A_{C 0 60})^{0.71}$  for PWR and

 $A_{C 14} = 0.02 \cdot (A_{C 0 60})^{0.76}$  for BWR.

and is presented in the diagrams. It can be seen that the correlations are of only average quality, as the correlation coefficients r = 0.85 (PWR) and r = 0.67 (BWR) are rather small and the scattering of the data from Japan and USA is quite large. The form of the functional dependence: A<sub>C 14</sub>  $\propto$  (A<sub>C0 60</sub>)<sup>0.71...0.76</sup> is nearly the same for PWR and BWR.

The country specific correlation analyses are shown in the following table. It can be seen that for the PWR data the correlations (Germany, USA) are better than for the BWR data (USA, Japan). The very good quality of the correlation for German BWR data must be interpreted in relation to the small number of data points.

Reactor	Country	ेक्ट्रॉ <b>a</b> r 5,771	<u>,</u>	`_` <b>r</b>	S	,, <b>F</b> ′,	n	Ac 14.= 522 2 10.00
PWR	Germany	0.75 ± 0.1	$-1.3 \pm 0.5$	0.79	0.85	46	29	$0.05 \cdot (A_{Co\ 60})^{0.75}$
	USA	$0.69 \pm 0.04$	$-0.24 \pm 0.2$	0.87	0.83	330	107	0.6∙(A <sub>Co 60</sub> ) <sup>0.69</sup>
1957 - 25 B 17	all	0.71 ± 0.04	$-0.53 \pm 0.2$	0.85	0.90	352	136	0.3·(A <sub>Co 60</sub> ) <sup>0.71</sup>
BWR	Germany	$0.87 \pm 0.15$	$-0.8 \pm 0.8$	0.96	1.16	33	5	0.16·(A <sub>Co 60</sub> ) <sup>0.87</sup>
	USA	$0.54 \pm 0.1$	$-0.9 \pm 0.4$	0.63	1.00	28	45	0.12·(A <sub>Co 60</sub> ) <sup>0.54</sup>
	Japan	$0.75 \pm 0.11$	$-1.8 \pm 0.3$	0.56	1.7	49	109	$0.02 \cdot (A_{Co\ 60})^{0.75}$
	all	0.76 ± 0.07	$-1.8 \pm 0.2$	0.67	1:53	123	159	$0.02 \cdot (A_{Co\ 60})^{0.76}$
a = slope, $b = y$ intersection, $r = correlation coefficient$ , $s = standard deviation$ , $F = F$ -test, $n = number of data points$								

### 4. Discussion

This result is consistent with the findings in [NUR 85] where also a generic scaling factor both for PWR and BWR was suggested. For PWR, a scaling factor of 0.0162 and for BWR, a scaling factor of 0.14 was calculated which is in good agreement with the values calculated here. In [NUR 85] it is expected that generic scaling factors may apply. This would not be in contradiction with the findings in this study, but it cannot be decided because of the broad prediction intervals.

The difference in the values of the pre-factors (0.3 for PWR and 0.02 for BWR) may be due to differences in the water chemistry. In PWRs the water is usually kept slightly basic (see main report, water chemistry) which can lead to the formation of insoluble carbonates. This mechanism could also explain why the pre-factor for PWR is about ten times the value for BWR. For BWRs, the water chemistry is substantially different, which may lead to the observed great variability in the data.

The prediction interval for the BWR data is quite large. The calculation of a generic scaling factor therefore seems not reasonable. This is true also if the correlation is evaluated only with data from single countries (see table above and [JAP 89] [NUR 85]).





The prediction interval for the PWR data is smaller and the quality of the correlation is better (r, F-value) than for the BWR data. Together with the knowledge about the water chemistry of C in PWR, the use of a generic scaling factor for PWRs seems possible.

From the data it cannot be concluded how e.g. differences in the water chemistry or variations in plant design between various countries influence C 14 contents in waste streams. This would require analyses for single plants for which the overall number of data would have to be considerably higher.

Correlation between C 14 and Co 60, PWR



C 14

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Correlation between C 14 and Co 60, BWR

7 ·

5 -

3

1

-1 -

-3 -

-5 -

log(C 14 [Bq/g])





<u>Cl 36</u>

# 1. Properties and Formation

Т <sub>й</sub>	3,01·10 <sup>5</sup> a					
Decay mode:		$\beta^{-}: \frac{36}{17}Cl \xrightarrow{\beta^{-}} \frac{36}{18}Ar$				
	EC, $\beta^+$ : ${}^{36}_{17}Cl \xrightarrow{\beta^+} {}^{36}_{16}S$					
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	<b>y(i)</b> E(i)			
β	9.81E-01	2.788E-01	2.74E-01			

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

Cl 36 is formed by thermal neutrons from Cl 35. A formation from K 39 and Cl 37 by fast neutrons is possible, but of minor relevance. Cl can be present as an impurity in the fuel. Sources for Cl 36 are activation of Cl 35 (chloride) in the coolant and activation of Cl 37 by fast neutrons in the fuel. Cl can also be contained in alloys e.g. in Inconel or borosilicate glasses.

# 2. Data and Correlation

No data for Cl 36 are available. For example, in Germany only very few samples have been analyzed for Cl 36, but only values below the detection limit have been found. If these data are however taken as real measurements and analyzed as the data for all the other nuclides (see diagram), it is obvious, that no correlation can be derived from this.

## 3. Discussion

It has been discussed [WTI 94b] that there is a possibility to assess the contents of Cl 36 via the contents of Na 22. The assumption is made that most of the Cl in the primary water should result from NaCl contents in water. Therefore, an assay of the Cl contents might be possible via the measurement of Na 22. This may be a promising approach which, however, has to be investigated separately. From the available data it is not possible to perform this kind of analysis.

Cl 36 - 1
Correlation between Cl 36 and Co 60, PWR





## 1. Properties and Formation

$\mathcal{T}_{\mathcal{H}}^{(n)}$ , where $\mathbf{T}_{\mathcal{H}}$ is a substitution of $\mathcal{T}_{\mathcal{H}}$ is a substitution of $\mathcal{T}_{\mathcal{H}}$ .	$1.4 \cdot 10^5$ a						
Decay mode:	EC: ${}^{41}_{20}Ca \xrightarrow{EC} {}^{41}_{19}K$						
Radiation	y(i) [Bq·s] <sup>-1</sup> E(i) [MeV] y(i) -						
Kα <sub>1</sub> X-ray	7.47E-02	3.314E-03	2.48E-04				
Kα <sub>2</sub> X-ray	3.75E-02	3.311E-03	1.24E-04				
Auger-KLL	6.78E-01	2.941E-03	1.99E-03				
Auger-KLX	9.88E-02	3.266E-03	3.23E-04				

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

Ca 41 is formed from Ca 40 by irradiation with thermal neutrons and from Ca 42 by irradiation with fast neutrons. It can also be a fission product from uranium, but this formation pathway will contribute only insignificantly.

Ca 41 is contained in concrete (large amounts of concrete arise from decommissioning of nuclear power plants, smaller amounts may arise from reconstruction work). Ca 41 is also contained in borosilicate glasses from core components and in zirconium alloys.

#### 2. Measurement

For a direct measurement, Ca 41 has first to be chemically separated and can then be counted with a liquid scintillation spectrometer.

A procedure for measurement of Ca 41 from concrete samples is described in [TÜV 93] as follows: After crushing, the initial (or start) procedure for concrete samples involves fusion with an equimolar mixture of sodium and potassium carbonates in a platinum crucible at about 1000 °C. Usually an excess of alkali metal carbonate of 6:1 is applied. The solidified melt is finely ground and leached with water until all sulfate is removed (BaCl<sub>2</sub>-Test). The insoluble residue is treated with hydrochloric acid (HCl:H<sub>2</sub>O = 1:1) and evaporated to dryness to precipitate silic acid completely. Next earth alkali metals are precipitated from acid solution as oxalates. Under these conditions (pH = 4.5) iron forms oxalato complexes and remains in solution. Earth alkali oxalates are destroyed at T = 900 °C to form oxides. After precipitation of earth alkali metal carbonates from slightly acidic solution barium is separated as barium chromate. Other trace contaminants are removed by scavenger precipitation using iron trihydroxide. From the filtrate calcium is isolated as calcium carbonate, and finally, redissolved in dilute hydrochloric acid.

#### 3. Data and Correlation

For Ca 41 there are only three data points available from Germany [TÜV 93], which are taken from activated concrete samples from nuclear power plants. As this does not allow to perform a full correlation analysis, only a scaling factor with Co 60 can be calculated:

Ca 41	Co 60		Ca 41/Co 60
124	371	Bq/g	0,33
126	576	Bq/g	0,22
37	103	Bq/g	0,36
		Mean:	0,30
		α:	0,07

As a first approach, a value of 0.3 can be assumed for a scaling factor between Ca 41 and Co 60 in concrete.

#### 4. \_\_\_\_ Discussion

Ca 41 is a nuclide that will become relevant during decommissioning of nuclear power plants but that is rarely measured for wastes from the operating phase. It has not been reported for the data sets from Sweden, Germany, USA or Japan. The scaling factor calculated here has to be used with caution as the data base is too small and can serve only as an orientation value which has to be backed up with future measurements.



## 1. Properties and Formation

	75000 a							
Decay mode:	Electron capture: ${}^{59}_{28}Ni \longrightarrow {}^{59}_{27}Co$							
Radiation	y(i) [(Bq:s) <sup>-1</sup> ]	E(i) [MeV]	y(i) E(i)					
Kα <sub>1</sub> X-ray	2.00E-01	6.930E-03	1.39E-03					
Kα <sub>2</sub> X-ray	1.02E-01	6.915E-03	7.02E-04					
Kβ <sub>1</sub> X-ray	2.71E-02	7.649E-03	2.08E-04					
Kβ3 X-ray	1.38E-02	7.649E-03	1.05E-04					

 $(y(i), E(i), y(i) \cdot E(i))$ : see beginning of appendix D for explanation)

Ni 59 is formed from Ni 58 by irradiation with thermal neutrons and from Ni 60 by irradiation with fast neutrons. Ni is usually contained in steels. Sources for Ni 59 are austenitic steel in the reactor and activation of Ni that is dissolved in the coolant and of corrosion particles deposited in the core (crud).

### 2. Measurement

An analytical technique for the measurement of Ni 59 and Ni 63 is described in [NUR 85]:

Solid samples such as sludge and resins are dissolved with mixed strong acids. Aliquots of the dissolved solids or of liquid samples are taken for analysis. Nickel is separated from contaminating nuclides by precipitating  $Fe(OH)_3$  with NH<sub>4</sub>OH. Nickel remains in solution as an ammonia complex. Further decontamination is accomplished by specific precipitation of Ni with dimethyl-glyoxine (Ni-DMG). The complex is dissolved and the organic destroyed with 1:1 HNO<sub>3</sub>:HClO<sub>4</sub> and heating to dryness. The salts are dissolved and volumetrically diluted. Yield is determined by atomic absorption. The Ni 63 activity is determined by liquid scintillation counting; the Ni 59 activity is measured using a thin-window NaI(Tl) detector.

## 3. Data and Correlation

Data are available from Sweden and Japan (for Japan only in the form of a correlation with Ni 63). Therefore, at first a correlation analysis between Ni 59 and Ni 63 is carried out. The statistical data show that the correlation is of good quality (r=0.94) and significant (F=251). Apparently, no distinction between BWR and PWR need to be made. The dependence is of the form

#### $A_{\text{Ni} 59} = 0.008 \cdot (A_{\text{Ni} 63})^{1.02}$

The slope of the curve is close to 1. It can be seen, however, that the activity ratios Ni 59/Ni 63 for the Swedish NPPs are smaller than the ones for Japanese NPPs. The following table shows the country specific results for the correlation between Ni 59 and Ni 63. Both the correlation for Swedish and Japanese data are very good (high r value) and show a slope near 1.

Reactor	Country	a	b	r	S	$\mathbf{F}_{\mathbf{x}}$	n	A <sub>Ni 59</sub> =
all	Sweden	$0.92 \pm 0.1$	$-3.0 \pm 0.1$	0.97	0.09	62	6	$0.001 \cdot (A_{Ni 63})^{0.92}$
Ni 63	Japan	$1.01 \pm 0.05$	$-1.8 \pm 0.1$	0.96	0.66	342	29	$0.02 \cdot (A_{Ni \ 63})^{1.01}$
· · · ·	all	1.02 ± 0.06	-2.1 ± 0.15	0.94	0.77	251	35	$0.008 \cdot (A_{Ni \ 63})^{1.02}$
a = slope, $b = y$ intersection $r = correlation coefficient, s = standard deviation, F = F-test, n = number of data points$								

A few additional data are available for a correlation of Ni 59 with Co 60 from Swedish PWR. Therefore, a second direct correlation is performed between for Ni 59 and Co 60. It can be seen that the correlation between Ni 59 and Co 60, for which only 6 data points are available is of poor quality, and the dependence

 $A_{Ni59} = 0.004 \cdot (A_{Co60})^{0.41}$ 

cannot be easily understood.

As an alternative for the direct correlation between Ni 59 and Co 60, this correlation can also be calculated from the correlation between Ni 59 and Ni 63 and between Ni 63 and Co 60 (see chapter for Ni 63):

 $A_{Ni 59} = 0.004 \cdot (A_{Co 60})^{0.94}$  for PWR  $A_{Ni 59} = 0.0008 \cdot (A_{Co 60})^{0.94}$  for BWR

#### 4. Discussion

The correlation between Ni 59 and Ni 63 is of very good quality. The data suggest that this correlation is valid on a generic basis. The slope of the correlation line is very close to 1 which is to be expected as the influence of the water chemistry should be the same for both Ni isotopes.

For PWR, a comparison of the results gained from the direct correlation with Co 60 and from the calculation via the two correlations Ni 59-Ni 63 and Ni 63-Co 60 shows that the scaling factor is the same but the exponent differs considerably. This suggests that the correlation Ni 59-Co 60 is not meaningful. It seems that a correlation of Ni 59 with Ni 63 is a better approach than a direct correlation of Ni 59 with Co 60. This, however, may be due to the fact that in total only very few data points have been available for this analysis.

Correlation between Ni 59 and Co 60





Ni 59

Correlation between Ni 59 and Ni 63



Ni 59

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# <u>Ni 63</u>

### 1. Properties and Formation

Ту		96 a	· · · · · · · · · · · · · · · · · · ·	
Decay mode:		$\beta^{-}: {}^{63}_{28}Ni \xrightarrow{\beta^{-}}{}^{63}_{29}Cu$		
Radiation	y(i) [(Bq-s) <sup>-1</sup> ]	E(i) [MeV]	y(i) · E(i)	• * ```
β <sup>-</sup>	1	1.713E-02	1.71E-02	

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

Ni 63 is formed from Ni 62 by irradiation with thermal neutrons, also from Zn 66 and Cu 63 by fast neutrons. Ni 63 contributes to the total activity after the decay of Co 60 and Cs 137. Sources for Ni 63 are austenitic steels in the core region, activation of dissolved Ni 62 in the coolant and of corrosion particles deposited in the core.

### 2. Measurement

Measurement of Ni 63 is possible after chemical separation with a liquid scintillation spectrometer.

An analytical technique for the separation and measurement of Ni 59 and Ni 63 is described in [NUR 85] (see chapter on Ni 59).

A procedure for the chemical separation of Fe and Ni in steel is described in [TÜV 93] as follows:

Metals and many alloys including steel are oxidized by mixtures of nitric and hydrochloric acid or nitric and hydrofluoric acid. If necessary, carriers for nickel and cobalt have to be added. The total amount of iron and nickel is determined photometrically. Next iron and nickel are coprecipitated from dilute acidic solution as hydroxides using sodium hydroxide. After dissolution in hydrochloric acid, nickel is separated from iron by excess ammonia. Under these conditions iron precipitates as iron trihydroxide [Fe(OH)<sub>3</sub>], whereas nickel remains in solution as [Ni(NH<sub>3</sub>)<sub>4</sub>]<sup>2+</sup>. This supernatant or filtrate containing nickel is further purified by the separation of bulk cobalt as tripotassium-hexa-nitrito-cobaltate, K<sub>3</sub>[Co(NO<sub>2</sub>)<sub>6</sub>], followed by the removal of traces of cobalt by anion exchange on Dowex 1-X8. Nickel is eluted by water. After precipitation of lanthanum hydroxide, nickel is separated from other radioisotopes by extracting the metal-organic complex of nickel dimethyl-glyoxime into chloroform. Next, chemical yield is determined, e.g. gravimetrically as the above mentioned complex or by AAS. The nickel organic complex is destroyed by nitric acid and the residue is taken up in hydrochloric acid. Finally, liquid scintillation counting is

performed. It is recommended to check the radiochemical purity with respect to remaining tracecontaminants by Ge/Li measurements.

A further method for a routinely measurement of Ni 63 (and Fe 55) after chemical separation is described in [ASS 94]. The procedure allows determination of the Ni 63 and Fe 55 contents in activated steels from the research reactor DIDO in order to clear them for controlled recycling and avoiding radioactive waste. Steel samples are subjected to a wet chemical process, and the low-energetic X-rays of Fe 55 (5.9 keV) and the  $\beta_{max}$ -energy (67 keV) of Ni 63 are measured in a liquid scintillation counter. The counting efficiency and the peak position in the spectrum is determined in a separate measurement by adding a specific amount of Fe 55 or Ni 63, respectively. From a series of measurements, the ratio Ni 63/Co 60 could be determined to 0.009. For Fe 55 and Ni 63, the lower limit of detection was determined to 2 Bq/g.

Additional information is contained in [ROO 94].

#### 3. Data and Correlation

Ni and Co are both commonly present in steels. Both elements are relatively insoluble or colloidal in a chemically neutral aqueous medium. Since their production and transport properties are similar, they can be expected to correlate well [NUR 85].

Data are available both for BWR and PWR for Sweden, Germany, USA and Japan. The correlation is performed with Co 60. For Japan, only measurements from NPPs with normal Co contents in steels have been included.

The correlations are of the form:

 $A_{Ni 63} = 0.47 \cdot (A_{Co 60})^{0.92}$  for PWR and  $A_{Ni 63} = 0.09 \cdot (A_{Co 60})^{0.92}$  for BWR.

It can be seen that the correlations are in both cases of good quality, as the correlation coefficients are near 1 and the scattering of the data is rather small. The country specific results in the following table show that the correlations are of rather good quality especially for data from Germany and USA, but also for BWR data from Japan. Swedish data do not correlate so well, but this must not be over-emphasised as the Swedish data cover only a comparatively small region at the lower end of the activity range.

PWR data sets from Sweden, Germany and USA result in nearly the same exponents a = 0.95 - 0.97, while Japanese data show a significantly smaller exponent value. The pre-factors, b, are also close together (0.2 - 0.8). BWR data sets, however, show exponent values ranging over a large interval (0.54 - 1.18) and also scattering pre-factors.



Reactor	Country	a	Ь	r	S ,	F	n	A <sub>Ni 63</sub> =
PWR	Sweden	$0.95 \pm 0.2$	$-0.1 \pm 0.2$	0.86	0.43	30	12	$= 0.8 \cdot (A_{Co \ 60})^{0.95}$
Co 60	Germany	0.97 ± 0.04	-0.66 ± 0.2	0.93	0.71	528	79	$= 0.2 \cdot (A_{Co \ 60})^{0.97}$
_ · · ·	USA	$0.95 \pm 0.03$	$-0.3 \pm 0.1$	0.95	0.68	974	104	$= 0.5 \cdot (A_{Co \ 60})^{0.95}$
	Japan	$0.82 \pm 0.05$	$-0.2 \pm 0.1$	0.88	0.68	233	68	$= 0.6 \cdot (A_{\text{Co} 60})^{0.82}$
	all	0.92 ± 0.02	$-0.33 \pm 0.07$	0 <b>.9</b> 4	0.69	2168	263	$= 0.47 \cdot (A_{Co\ 60})^{0.92}$
BWR	Sweden	$0.54 \pm 0.07$	$-1.06 \pm 0.07$	0.80	0.35	55	34	$= 0.09 \cdot (A_{Co \ 60})^{0.54}$
Co 60	Germany	$0.92 \pm 0.05$	-0.67 ± 0.2	0.90	0.75	320	75	$= 0.21 \cdot (A_{Co60})^{0.92}$
	USA	0.97 ± 0.04	$-1.6 \pm 0.2$	0.93	0.63	518	79	$= 0.03 \cdot (A_{Co60})^{0.97}$
in a start and a start	Japan	$1.18 \pm 0.08$	$-1.3 \pm 0.2$	0.96	0.55	202	20	$= 0.05 \cdot (A_{Co  60})^{1.18}$
	all and a		-1.04 ± 0.09	0.94	0.72	1623	s. ; <b>20</b> 8	$= 0.09 \cdot (A_{Co60})^{0.92}$
a = slope,	b = y inters	ection, $\mathbf{r} = correl$	ation coefficient	, s = stand	ard devia	tion, F = F	-test, n =	number of data points

#### 4. Discussion

The form of the functional dependence:  $A_{Ni 63} \propto (A_{Co 60})^{0.92}$  is the same for PWR and BWR, only the pre-factors differ. The deviation of the exponent (0.92) from 1 seems to be significant because of the small error range (± 0.02). Reasons for this could be e.g. differences of filter efficiencies for Ni 63 and Co 60. However, the country specific analyses (especially PWR data from Sweden, Germany and USA as well as BWR data from Germany and USA) would also be in agreement with an exponent a near or equal to 1.

The rather small scattering of the data and the country specific analyses suggest that no distinction need to be made between the data from various countries. This result is consistent with the findings in [NUR 85] where it was suggested that generic scaling factors for LWR may apply. The scaling factor for PWR was found to be approximately 6 times the one for BWR (which is in agreement with the results of this study). In [NUR 85] this is explained with the increased amounts of Co 60 in BWR systems from the stellite material<sup>1</sup> used on control blades.

It can be concluded that generic scaling factors exist for PWR and BWR.

1)

#### Correlation between Ni 63 and Co 60, PWR



Correlation between Ni 63 and Co 60, BWR







# 1. Properties and Formation

Tig		29.12 a	
Decay mode:		$\beta^{-}: {}^{90}_{38}Sr \xrightarrow{\beta^{-}} {}^{90}_{39}Y$	
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	y(i) E(i)
β-	l	1.957E-01	1.96E-01

 $(y(i), E(i), y(i) \cdot E(i):$  see beginning of appendix D for explanation)

Sr 90 is a fission product from U. Sr 90 can also be formed by activation of zirconium in steel or zircaloy, but to a lesser extent. The main source of Sr 90 are the fuel elements.

## 2. Measurement

An analytical technique for the measurement of Sr 89 and Sr 90 is described in [NUR 85]:

Solid samples such as sludge and resins are dissolved or leached with mixed strong acids. Aliquots of the dissolved solid or liquid samples are taken for analysis. A combination of precipitation and scavenging techniques are used for decontamination. Yttrium is separated and strontium is precipitated as the carbonate and counted. A two-week yttrium in-growth period is allowed; the carbonate is dissolved and the yttrium is separated, purified, and counted. The Sr 90 concentration is derived from Y 90 counting. Sr 89 concentration is calculated by subtracting Sr 90 and Y 90 contributions to the counting rate of the strontium carbonate. For old samples that contain no Sr 89, the first count of the strontium activity ends the analysis. A gas proportional counter is required for counting.

# 3. Data and Correlation

In [NUR 85], it is pointed out that Sr exists primarily in solution as ions rather than as particulates and that in a PWR with its sometimes slightly basic chemistry, some of the Sr may be in an insoluble form such as carbonate. It is estimated that Cs is the only candidate for a correlation, although Sr and Cs have different collection efficiencies on filters or ion-exchange beds and decontamination factors for Cs across resin beds are often low and variable compared to Sr.

Data are available both for PWR and BWR for Sweden, Germany, USA and Japan. The correlations are of the form:

 $A_{Sr 90} = 0.01 \cdot (A_{Cs 137})^{0.90}$  for PWR and

## $A_{Sr 90} = 0.07 \cdot (A_{Cs 137})^{0.85}$ for BWR

and are shown in the diagrams. The country specific analysis in the following table reveals that the exponents vary considerably for data sets from various countries. PWR data from Germany, USA and Japan show exponent values rather close together (a = 0.81 - 0.85), while Swedish data show a higher exponent value. The reason for this is (like for Ni 63, see above) that the Swedish data are not distributed homogeneously over the activity range (see PWR diagram) but form two clusters in the low and the high activity region (if Swedish data only from the low activity cluster are analysed, the exponent is near 0.6). Pre-factors lie close together.

For BWR data, exponents have similar values as for PWR data (a = 0.7 - 0.83). The German BWR data that result in deviating values for exponent and pre-factor must not be over-emphasised as the data set is small (n = 8), compared with 84 and 99 data points for USA and Japan.

Reactor	Country	à â	b	r	S	F	<b>n</b>	$A_{Sr90} =$
PWR	Sweden	$1.13 \pm 0.05$	$-1.6 \pm 0.1$	0.97	0.62	501	35	$0.03 \cdot (A_{Cs \ 137})^{1.13}$
	Germany	$0.83 \pm 0.1$	$-1.5 \pm 0.6$	0.85	0.74	54	23	$0.03 \cdot (A_{Cs \ 137})^{0.83}$
	USA	$0.85 \pm 0.05$	$-1.8 \pm 0.2$	0.85	1.18	261	101	$0.02 \cdot (A_{Cs \ 137})^{0.85}$
	Japan	$0.81 \pm 0.06$	$-1.9 \pm 0.1$	0.87	0.86	162	54	$0.01 \cdot (A_{Cs \ 137})^{0.81}$
· ·	all	$0.90 \pm 0.03$	$-1.95 \pm 0.09$	0.93	1.01	1259	213	$0.01 \cdot (A_{Cs \ 137})^{0.90}$
BWR	Germany	$0.7 \pm 0.1$	$-0.4 \pm 0.7$	0.88	1.2	21	8	$0.4 \cdot (A_{Cs \ 137})^{0.7}$
· · · ·	USA	$0.83 \pm 0.04$	$-1.0 \pm 0.2$	0.92	0.81	445	84	$0.1 \cdot (A_{Cs \ 137})^{0.83}$
	Japan	$0.81 \pm 0.04$	$-1.2 \pm 0.1$	0.91	0.87	452	99	$0.06 \cdot (A_{Cs \ 137})^{0.81}$
· · · ·	all	0.85 ± 0.02	-1.13 ± 0.07	0.95	0.88	1735	193	$0.07 \cdot (A_{Cs \ 137})^{0.85}$
a = slope,	a = slope, b = y intersection, r = correlation coefficient, s = standard deviation, $F = F$ -test, n = number of data points							

#### 4. Discussion

It can be seen that the correlations are in both cases of rather good quality, as the correlation coefficients r = 0.93...0.95, which is also supported by the high F values. In the PWR case, the scattering of data seems to be slightly larger than for BWR. The form of the functional dependence:  $A_{Sr 90} \propto (A_{Cs 137})^{0.85..0.90}$  is similar for PWR and BWR, only the pre-factors differ.

However, as was pointed out in [NUR 85], the nuclides of Sr and Cs behave differently in filters and ion exchange resins. Therefore it is recommended to perform a correlation between Sr 90 and Cs 137 on a plant specific basis. Despite the great number of samples investigated in [NUR 85], it was stated that the data basis was not sufficient for a both plant and waste stream specific analysis.

In contrast to the view expressed in [NUR 85], it has been found here that data from all four countries show similar results as discussed above. The conclusion must be drawn, that a generic correlation analysis provides meaningful results.

Correlation between Sr 90 and Cs 137, PWR



Correlation between Sr 90 and Cs 137, BWR





## 1. Properties and Formation

T <sub>K</sub>	3500 a						
Decay mode:	Electron capture: ${}^{93}_{42}Mo \xrightarrow{EC} {}^{93}_{41}Nb$						
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	y(i) · E(i)				
Kα <sub>1</sub> X-ray	3.45E-01	1.662E-02	5.73E-03				
Kα₂ X-ray	1.81E-01	1.652E-02	2.99E-03				
Kβ1 X-ray	5.76E-02	1.862E-02	1.07E-03				

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

Mo 93 is formed from Mo 92 by irradiation with thermal neutrons. In addition, formation from Ru 96 and Mo 94 by fast neutrons is possible, but to a lesser extent. Ruthenium and molybdenum are present as impurities in  $UO_2$ . Molybdenum can be present as alloy element in steels and in lubricants. Sources for Mo 93 are metallic materials, esp. steels and zirkaloy in the core region, e.g. from the reactor pressure vessel, and activation of molybdenum in dissolved form or as corrosion particles in the coolant.

### 2. Data and Correlation

Mo and Co are both present in steels as alloy elements. The abundance of Mo with respect to Co varies considerably with the kind of steel (the ratio of Ni to Co, for example, varies less). All the same, a correlation with Co is to be expected.

No data are available for Mo 93.



## <u>Tc 99</u>

### 1. Properties and Formation

T <sub>1/2</sub>		213000 a				
Decay mode:	$\beta^{-}: \stackrel{99}{_{43}}Tc \xrightarrow{\beta^{-}} \stackrel{99}{_{44}}Ru$					
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	y(i) · E(i)			
β <sup>-</sup>	1	1.013E-01	1.01E-01			

Tc 99 is formed by irradiation of Mo 98 with thermal neutrons. If Tc 99 is present as a fission product, the irradiation process is of minor importance. Sources for Tc 99 are uranium fission in the fuel and molybdenum as an alloy element in steels and zircaloy, but with minor importance.

### 2. Measurement

An analytical technique for the measurement of Tc 99 is described in [NUR 85] as follows:

Rhenium is substituted for technetium as a carrier. Aliquots of liquid samples are used for analysis. Solid samples are dissolved by alkaline fusion to prevent volatilisation of technetium. Subsequent solutions are treated with 8 M HNO<sub>3</sub> and boiled to remove iodine. Cobalt is repeatedly used as a scavenging agent to remove radiocobalt and other transition metal radionuclides. The Re(Te) is precipitated as a complex with tetraphenyl arsonium chloride and the Tc 99 activity is determined by gas proportional counting.

### 3. Data and Correlation

Because Tc 99 is a fission product, a correlation with Cs 137 should be expected. In [NUR 85] it is stated that Tc 99 is soluble in coolant but that the removal of Tc 99 by radwaste systems is not well understood. Therefore, the same cautious approach towards generic scaling factors applies as for Tc 99 (see Sr 90) so that scaling factors should best be determined on a plant and waste stream specific basis.

For Sweden and USA, data are available both for PWR and BWR. Data from Germany is only available from BWR, no data is available from Japan. The correlations are of the form:

 $A_{Tc 99} = 5 \cdot 10^{-4} \cdot (A_{Cs 137})^{0.81}$  for PWR and

 $A_{Tc 99} = 2 \cdot 10^{-4} \cdot (A_{Cs 137})^{0.86}$  for BWR

and are shown in the diagrams. The country specific analyses (following table) demonstrate, however, that the overall correlations must be regarded with caution as the data sets for Sweden (PWR and BWR) and USA (BWR) show almost no correlation (small r and F values, large error ranges for a). The reason that the complete data sets (data of all countries) correlate with acceptable quality, while the country specific data sets do not correlate or only with poor quality, is the fact that the Swedish data cover the low activity region while the USA data fall mainly in the high activity region (cf. diagrams). If both data sets are combined, the resulting data set extends over the whole activity range and produces a correlation.

Reactor	Country	a	<b>b</b>	r .	` S	F ·	.n ´	$A_{Tc 99} =$
PWR	Sweden	$0.16 \pm 0.15$	$-3.0 \pm 0.2$	0.32	0.3	1	11	no correlation
	USA	$0.62 \pm 0.15$	$-2.2 \pm 0.8$	0.74	0.92	17	16	$0.006 \cdot (A_{Cs \ 137})^{0.6}$
	all	$0.81 \pm 0.06$	$-3.3 \pm 0.3$	0.94	0.80	183	<b>27</b>	$0.0005 \cdot (A_{Cs \ 137})^{0.81}$
BWR	Sweden	$0.35 \pm 0.15$	$-3.3 \pm 0.1$	0.43	0.62	6	29	no correlation
•	USA	$0.4 \pm 0.4$	$-0.7 \pm 2$	0.44	1.1	1	7	no correlation
	all	0.86 ± 0.06	$-3.7 \pm 0.2$	0.92	0.84	185	37	$0.0002 \cdot (A_{Cs \ 137})^{0.86}$
a = slope.	b = v inters	ection, $r = correl$	ation coefficient	s = stand	lard devia	tion. $F = F$	-test. n = 1	number of data points

#### 4. Discussion

The correlations of the whole data sets are in both cases of satisfactory quality, as the correlation coefficients r = 0.92...0.94. The scattering of data is rather large, which is also reflected by the only average values of F. The form of the functional dependence:  $A_{Tc 99} \propto (A_{Cs 137})^{0.81..0.86}$  is almost the same for PWR and BWR with also the same pre-factors. This must, however, be interpreted cautiously as the country specific data sets do not correlate well as was outlined above. Results of this study do not allow to perform a generic correlation for PWR or BWR.

This is supported by [NUR 85], where different behaviour of Tc and Cs in the radwaste systems is described, and it is therefore recommended to perform a correlation between Tc 99 and Cs 137 only on a plant specific basis. The number of samples investigated in [NUR 85] was however too small for a both plant and waste stream specific analysis.

Correlation between Tc 99 and Cs 137, PWR





Tc 99

Correlation between Tc 99 and Cs 137, BWR





## 1. Properties and Formation

T <sub>iź</sub>	1,57·10 <sup>7</sup> a						
Décay mode:	$\beta^{-}: \stackrel{129}{_{53}}I \xrightarrow{\beta^{-}} \stackrel{129}{_{54}}Xe$						
Radiation	$\overline{y}(i) [(Bq \cdot s)^{-1}] = E(i) [MeV] $ $y(i) \cdot E(i)$						
β <sup>-</sup>	1.0	4.888E-02	4.89E-02				
γ	7.51E-02	3.958E-02	2.97E-03				
Kα <sub>I</sub> X-ray	3.69E-01	2.978E-02	1.10E-02				
Kα <sub>2</sub> X-ray	1.99E-01	2.946E-02	5.87E-03				

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

I 129 is mainly a fission product from U. The formation from Te 128 by thermal neutrons and from Xe 129 with fast neutrons is of minor importance, as these elements are usually not present in construction materials in nuclear power plants. Sources for I 129 are the fuel elements and uranium material impurities.

### 2. Measurement

An analytical technique for the measurement of I 129 is described in [NUR 85] as follows:

Aliquots of liquid samples are used for analysis. Solid samples are dissolved in alkaline fusion to prevent volatilization of iodine. Subsequent solutions are stirred with anion exchange resin in batch form to remove iodine. The iodine is extracted from the resin and liquid-liquid extractions are performed. The purified iodine is precipitated as CuI and counted on a thin-window NaI(Tl) detector.

### 3. Data and Correlation

For I 129, German data are only available for BWR. For USA and Japan data are available both for PWR and BWR.

The correlations are of the form:

 $A_{1129} = 0,02 \cdot (A_{Cs 137})^{0.3}$  for PWR,

 $A_{1129} = 8 \cdot 10^{-6} \cdot (A_{Cs \ 137})^{0.9}$  for BWR, all countries and  $A_{1129} = 8 \cdot 10^{-7} \cdot (A_{Cs \ 137})^{0.96}$  for BWR, Japan only.

It can be seen that the correlations are in the first two cases of poor quality, as the correlation coefficients r are only 0.5 for PWR and 0.7 for BWR. The scattering of data is rather large and the low value of F for PWR indicates that this correlation is not very significant. The country specific analysis in the following table shows that data from USA are not correlated (r is small for PWR and BWR data), while data from Japan, especially for BWR, are well correlated (high r and F values). In addition, the correlation for Japanese BWR data has an exponent is near 1. In conclusion, the correlation of the data sets must mainly be attributed to the Japanese data. The analysis of the well correlated Japanese data together with uncorrelated data from other countries (BWR, USA, Germany) will result in a correlation of poorer quality that is not significant.

Reactor	Country	a		r i r	S	F	n	A <sub>I 129</sub> =
PWR	USA	$0.1 \pm 0.1$	$-0.4 \pm 0.5$	0.22	0.73	0.6	15	no correlation
Cs 137	Japan	$0.6 \pm 0.2$	$-3.8 \pm 0.7$	0.83	0.77	7	5	$0.00016 \cdot (A_{C_{s 137}})^{0.6}$
	all	0.3 ± 0.1	$-1.8 \pm 0.6$	0.49	1.1	6	20	$0.02 \cdot (A_{Cs \ 137})^{0.3}$
BWR	USA	$0.1 \pm 0.2$	$0.0 \pm 0.9$	0.09	1.3	0.1	15	no correlation
Cs 137	Japan	$0.95 \pm 0.08$	$-6.1 \pm 0.3$	0.95	0.7	139	18	$8 \cdot 10^{-7} \cdot (A_{Cs137})^{0.95}$
	all	0.9 ± 0.15	-5.1 ± 0.6	0.73	. 1 <b>.81</b>	36	34	8·10 <sup>-6</sup> ·(A <sub>Cs137</sub> ) <sup>0.9</sup>
a = slope, $b = y$ intersection, $r = correlation$ coefficient, $s = standard$ deviation, $F = F$ -test, $n = number$ of data points								

#### 4. Discussion

According to [NUR 85], iodine and Cs have similar transport properties in reactor systems and release mechanisms from reactor fuel but filter systems have different retention properties for I and Cs. Cs and I might therefore show considerable scattering when being correlated. In [NUR 85] it was further suggested that different correlation behaviours may exist for the ranges of low and of high specific activities.

This is not in contradiction with this study. The results presented above show that no generic correlation is possible. In addition, the measurements suggest that Japanese data always tend to lower specific activity ratios I 129/Cs 137 compared to the measurements from the USA.

It must therefore be concluded that at least for PWR a correlation analysis must be performed on a plant specific basis, if it is intended at all. For BWR, the functional dependence of  $A_{I129} \propto (A_{Cs\,137})^{0.9..1}$  suggests that a real correlation may exist. However, this estimate is mainly based on the Japanese data.

In order to check whether the assumption of different correlation behaviours for lower and higher activity regions is correct, more data points would have to be analyzed.

Correlation between I 129 and Cs 137, PWR





1 129



Correlation between I 129 and Cs 137, BWR

영상품 김 사람들이 있는 것

Correlation between I 129 and Cs 137, BWR Japan





I 129



# <u>Cs 135</u>

## 1. Properties and Formation

$T_{t/2}$		2,3·10 <sup>6</sup> a	
Decay mode:		$\beta^{-}: \stackrel{135}{}_{55}Cs \xrightarrow{\beta^{-}} \stackrel{135}{}_{56}Ba$	
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	y(i) · E(i)
β <sup>-</sup>	1	6.733E-02	6.73E-02

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

Cs 135 is a fission product from uranium. Cs 135 is contained in fuel elements and in uranium material impurities.

### 2. Data and Correlation

Cs 135 and Cs 137 are both single neutron fission products. Therefore, a simple linear correlation between these nuclides should be expected.

No data are available for a correlation analysis. There are, however, several measurements of high-active waste in [DOE 92] that indicate that an activity ratio of Cs 135/Cs 137 according to the half-lives of the two nuclides can be expected in different waste streams. The following table shows data from several sites.

Cs 135 [Bq/package].	Cs 137 [Bq/package]	Activity ratio	Origin	Reférence [DOE 92]
2,35E+10	1,05E+15	2,2E-05	West Valley Demonstration Project	Tab. 3.2.3
3,68E+09	1,61E+15	2,3E-06	Savannah River Site	Tab. 3.3.3
9,29E+09	1,89E+15	4,9E-06	Hanford Site	Tab. 3.4.4
7,47E+09	1,34E+15	5,6E-06	Hanford Site	Tab. 3.4.5
3,45E+09	6,14E+14	5,6E-06	Idaho Nat. Engineering Laboratory	Tab. 3.5.3
	Average:	8,2E-06		

Cs 135 - 1

#### 3. Discussion

The average activity ratio between Cs 135 and Cs 137 of  $8 \cdot 10^{-6}$  is in correspondence with the ratio that is calculated from the half lives of the two nuclides  $(1,5 \cdot 10^{-5})$  under the assumption that the fission production rate of both nuclides is identical. As this together with similar chemical behaviour in the water system and therefore in the contamination can well be expected, it can be assumed that also in LWR waste a ratio of about  $10^{-5}$  between the activities of Cs 135 and Cs 137 applies. It must be noted, however, that the date given above are for HLW and for various types of plants so that application to normal wastes from light water reactors must be done with caution.



# <u>U 234</u>

### 1. Properties and Formation

T <sub>12</sub>		244500 a	
Decay mode:		$\alpha: \xrightarrow{234}{92} U \xrightarrow{\alpha} \xrightarrow{230}_{90} Th$	
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	<b>ý(i)</b> • <b>Ē(i)</b>
α	2.74E-01	4.721	1.30E+00
α	7.23E-01	4.773	3.45E+00

(y(i), E(i), y(i) · E(i): see beginning of appendix D for explanation)

U 234 is a decay product from  $U_{nat}$  238. It enters the contamination from the fuel elements and may also originate from U impurities in materials. The abundance of U 234 depends on the enrichment of the fuel.

#### 2. Measurement

An analytical technique for the separation and measurement of U 234 is described in [NUR 85] as follows:

The sample is dissolved by fusion technique. The fusion cake is dissolved in HCl and the transuranic nuclides are coprecipitated with barium sulfate. The barium sulfate is dissolved and liquidliquid extractions are performed separating U, Th; Pu, Np; and Am, Cm. Each fraction is electroplated and analyzed using a surface barrier alpha-particle detector. A gamma-ray analysis of the Am-Cm sample gives the Ce 144 in the sample. The nuclide Ce 141 is added to the initial sample as a tracer.

#### 3. Data and Correlation

As there is no formation pathway for U in the sense as for fission products or activated corrosion products, no correlation with Co 60 or Cs 137 can be expected. However, it may be reasonable to correlate U 234 to U 235 or U 238 and to analyse the U 235 or the total U contents in the waste. Data for U 234 are only available for German PWR.

From the diagram it is obvious that no correlation between U 234 and Cs 137 exists (note also the very low values of r and F). Not even an approach for a correlation on a phenomenological basis seems reasonable.

For the correlations with U 235 and U 238 the correlations are of the form

 $A_{U\,234} = 35 \cdot (A_{U\,235})^{1.24}$  and  $A_{U\,234} = 3.8 \cdot (A_{U\,238})^{1.13}$ 

#### 4. Discussion

From the two diagrams for U 234 with U 235 and U 238, respectively, it can be seen that the correlations are very good with correlation coefficients r=0.97..0.99. The F values indicate statistical significance.

It must be noticed that the exponents significantly deviate from 1.0 (1.24 for U 234-U 235 and 1.13 for U 234-U 238). From simple considerations, a linear interdependence should have been assumed at least with U 235 due to the enrichment process.

From [THI 94c] it can be concluded that no correlation between U 235 or U 238 and Cs 137 exists. That implies that the knowledge of the correlations between U 234 and the other U isotopes cannot be used to calculate the correlation with Cs 137.

For both correlations, a distinction between PWR and BWR is not necessary.

Correlation between U 234 and Cs 137





U 234

Correlation between U 234 and U 235



Correlation between U 234 and U 238





U 234



## <u>Np 237</u>

### 1. Properties and Formation

T <sub>14</sub>		2.14·10 <sup>6</sup> a	
Decay mode:		$\alpha: \stackrel{^{237}}{_{g_3}}Np \xrightarrow{\alpha} \stackrel{^{233}}{_{g_l}}Pa$	
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	y(i) · E(i)
α	6.19E-02	4.640	2.87E-01
α	2.50E-01	4.772	1.19E+00
α	4.71E-01	4.789	2.25E+00
γ	1.40E-01	2.937E-02	4.11E-03
γ	1.26E-01	8.650E-02	1.09E-02

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

Np 237 is present in irradiated uranium. Sources for Np 237 are the fuel elements and uranium material impurities.

#### 2. Measurement

An analytical technique for the separation and measurement of Np 237 is described in [NUR 85] (see U 234 chapter)

### 3. Data and Correlation

There are only 5 data points available from Sweden from which it is impossible to perform a meaningful correlation analysis. In addition, for these data points no measurements of Cs 137 have been reported, so a different key nuclide must be chosen. A tentative correlation is performed between Np 237 and U 238 which produces the best results in comparison to other TRU nuclides. A second correlation analysis is performed between Np 237 and U 235 for which only 4 data points are available.

It is evident that the correlations

 $A_{Np 237} = 29 \cdot (A_{U 238})^{0.84}$  and  $A_{Np 237} = 0.09 \cdot (A_{U 235})^{1.34}$ 

are of rather poor quality (large region of confidence, r and F small for U 238) and cannot serve as a basis for any prediction.

# 4. Discussion

•

From the data that are available no correlation analysis is possible. Further investigation is necessary for Np 237.

Correlation between Np 237 and U 235

log(Np 237 [Bq/g])


Correlation between Np 237 and U 238

1. S. S. 18 .

1 - C



Np 237



# 1. Properties and Formation

<b>T</b> 12	87.74 a							
Decay mode:	$\alpha: \xrightarrow{238}_{94} \mathrm{Pu} \xrightarrow{\alpha} \xrightarrow{234}_{92} \mathrm{U}, \mathrm{SF}$							
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	ý(i) • E(i)					
α <sub>15</sub>	2.83E-01	5.456E+00	1.54E+00					
α <sub>16</sub>	7.16E-01	5.499E+00	3.94E+00					

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

Pu 238 is formed by neutron reactions or during decay processes from uranium and transuranium isotopes. With thermal neutrons, Pu 238 is formed from Np 237. Pu 238 is contained in irradiated uranium. Sources for Pu 238 are the fuel elements and uranium material impurities.

## 2. Measurement

An analytical technique for the separation and measurement of Pu 238 is described in [NUR 85] (see U 234 chapter).

# 3. Data and Correlation

For Pu 238 and Pu 240, in [NUR 85] no direct correlation with Cs 137 was investigated but these nuclides were correlated to the total activity of Pu 239+Pu 240. For this activity in turn correlations were investigated with Ce 144, Cs 137 and Co 60. It was found that the correlation with Ce 144 was very satisfactory. This, however, would be a difficult approach in the present investigation as the number of available measurements that include Ce 144 is small. In addition, this nuclide is not reported for each country. So a direct correlation of Pu 238 with Cs 137 is used. Data are available from Sweden, Germany and USA. It must be emphasised that all data points represent actual measurements and do not result from calculations.

The resulting correlations are of the form

 $A_{Pu 238} = 3.7 \cdot 10^{-4} \cdot (A_{Cs 137})^{0.80}$  for PWR and  $A_{Pu 238} = 4.5 \cdot 10^{-3} \cdot (A_{Cs 137})^{0.65}$  for BWR

and are presented in the first and second diagram. The quality of these correlations is comparatively poor, as is indicated by the low values of r and the large scattering, especially in the BWR case, but they are statistically significant (F values 186 and 760). Slightly better results can be obtained from a correlation with Co 60. The resulting correlations are of the form:

 $A_{Pu\;238}=3.3{\cdot}10^{-6}{\cdot}(A_{Co\;60})^{1.07}$  for PWR and

 $A_{Pu 238} = 2.5 \cdot 10^{-5} \cdot (A_{Co 60})^{0.89}$  for BWR.

and are presented in the third and fourth diagram. The quality of these correlations is rather good with values of r = 0.92...0.96 and much higher F values.

Finally, an excellent correlation of Pu 238 is possible with Pu 239+Pu 240. The resulting correlation is of the form:

 $A_{Pu\ 238} = 1.57 \cdot A_{Pu\ 239/240}$ 

and is presented in the fifth diagram. The correlation is perfect (r = 1.0, very high F value) and no distinction need to be made between PWR and BWR.

The country specific analysis in the following table confirms these results. No country specific peculiarities are observed. For the PWR and BWR correlations with Co 60 it is important to note that the German and Swedish data sets cover different activity regions (for PWR: Sweden low activities, Germany higher activities, for BWR vice versa; cf. diagrams). Therefore, the resulting correlations of the combined data sets must be interpreted with caution. - The correlations with Pu 239/240 are identical for the Swedish and German data sets.

Reactor	Country	a	b b	; <b>r</b>	S	F	n	A <sub>Pu 238</sub> =
PWR	Sweden	$0.83 \pm 0.05$	$-4.3 \pm 0.1$	0.95	0.60	299	35	$5 \cdot 10^{-5} \cdot (A_{Cs \ 137})^{0.83}$
Cs 137	Germany	0.61 ± 0.07	$-2.7 \pm 0.3$	0.76	0.97	85	65	$2 \cdot 10^{-3} \cdot (A_{Cs \ 137})^{0.61}$
	USA	0.69 ± 0.05	$-2.9 \pm 0.2$	0.89	1.29	232	152	$1.3 \cdot 10^{-3} \cdot (A_{C_{s} \ 137})^{0.69}$
	all	0.80 ± 0.03	$-3.4 \pm 0.1$	0.87	1.23	759	252	$3.7 \cdot 10^{-4} \cdot (A_{Cs137})^{0.80}$
BWR	Germany	$0.7 \pm 0.15$	$-3.0 \pm 0.6$	0.61	1.41	17	30	$1 \cdot 10^{-3} \cdot (A_{Cs137})^{0.7}$
Cs 137	USA	$0.65 \pm 0.05$	$-2.2 \pm 0.2$	0.78	1.25	184	120	$6.3 \cdot 10^{-3} \cdot (A_{C_{s} 137})^{0.65}$
	all	0.65 ± 0.05	$-2.3 \pm 0.2$	0.74	1.31	183	151	$4.5 \cdot 10^{-3} \cdot (A_{Cs \ 137})^{0.65}$



Reactor	Country	а	b ्	<b>. r</b> .	S	F	n	$A_{Pu \ 238} =$	
PWR	Sweden	$1.01 \pm 0.04$	$-6.14 \pm 0.07$	0.98	0.38	768	38	$7.2 \cdot 10^{-7} \cdot (A_{Co60})^{1.01}$	
Co 60	Germany	$0.64 \pm 0.07$	$-3.3 \pm 0.3$	0.76	0.94	99	74	$5 \cdot 10^{-4} \cdot (A_{Co \ 60})^{0.64}$	
te da esta esta esta esta esta esta esta est	all	1.07 ± 0.04	∴-5.5 ± 0.2	0.92	1.12	587	_ 112	$3.3 \cdot 10^{-6} \cdot (A_{Co\ 60})^{1.07}$	
BWR	Sweden	$0.93 \pm 0.08$	$-5.1 \pm 0.9$	0.92	0.66	135	26	$8 \cdot 10^{-6} \cdot (A_{Co\ 60})^{0.93}$	
Co 60	Germany	$0.8 \pm 0.1$	$-4.3 \pm 0.5$	0.77	1.2	49	35	$5 \cdot 10^{-5} \cdot (A_{Co \ 60})^{0.8}$	
		0.89 ± 0.03	-4.6 ± 0.3	0.96	1.0	713	· 61	2.5.10 <sup>-5</sup> .(A <sub>Co 60</sub> ) <sup>0.89</sup>	
all	Sweden	1.003±0.008	$0.30 \pm 0.05$	1.00	0.46	12968	76	$2.0 \cdot (A_{Pu \ 239/240})^{1.00}$	
Pu 239/	Germany	$0.98 \pm 0.02$	$0.11 \pm 0.03$	0.98	0.28	3087	105	$1.3 \cdot (A_{Pu \ 239/240})^{0.98}$	
Pu 240	all	1.01 ± 0.01	0.20 ± 0.03	1.00	<b>.0.38</b>	21530	181	$1.57 \cdot (A_{Pu}_{239/240})^{1.01}$	
a = slope,	a = slope, b = y intersection, r = correlation coefficient, s = standard deviation, F = F-test, n = number of data points								

### 4. Discussion

The results obtained here from Swedish and German data is in very good agreement to US data [NUR 85], from which a scaling factor of 1.37 for Pu 238 with Pu 239/240 has been calculated. The difference may be explained by varying burn-up of the fuel on which the activity ratio of Pu 238 and Pu 239/240 depends. The exponent in this correlation is 1 which is in accordance to the fact that these nuclides have similar formation pathways.

It can be concluded that the best way to calculate correlations for Pu 238 is to use the correlation with Pu 239+Pu 240 and then to use the correlation between Pu 239+Pu 240 and Cs 137. A correlation with Co 60 seems to be possible, too, but the country specific results discussed above indicate that the results must be interpreted carefully. Due to the fact that no common formation pathways for both nuclides exist, such a correlation might better be performed on a plant specific basis.

A correlation of Pu 238 with Cs 137 does not seem promising. If it is tried all the same, a plant specific approach might be reasonable.

Correlation between Pu 238 and Cs 137, PWR







Correlation between Pu 238 and Cs 137, BWR

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AS CONT

Correlation between Pu 238 and Co 60, PWR







Correlation between Pu 238 and Co 60, BWR

1 **1** 5

Correlation between Pu 238 and Pu 239+Pu240





Pu 238

Pu 239/240 - 1



# <u>Pu 239+Pu 240</u>

# 1. Properties and Formation

### Pu 239:

Ту	24065 a							
Decay mode:	$\alpha: \xrightarrow{239}_{94} Pu \xrightarrow{\alpha} \xrightarrow{235}_{92} U$							
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	y(i) E(i)					
α	1.07E-01	5.105	5.45E-01					
α	1.52E-01	5.143	7.83E-01					
α	7.38E-01	5.156	3.80E+00					

 $(y(i), E(i), y(i) \cdot E(i)$ : see beginning of appendix D for explanation)

## Pu 240:

T <sub>12</sub>	6537 a							
Decay mode:	$\alpha: \frac{240}{94} Pu \xrightarrow{\alpha} \frac{236}{92} U$ , spontaneous fission							
Radiation	y(i) [(Bq:s) <sup>-1</sup> ]	E(i) [MeV]	y(i) · E(i)					
α	2.65E-01	5.124	1.36E+00					
α	7.34E-01	5.168	3.79E+00					

 $(y(i), E(i), y(i) \cdot E(i))$ : see beginning of appendix D for explanation)

Pu 239 and Pu 240 are contained in irradiated uranium. Sources for Pu 239 and Pu 240 are the fuel elements and uranium material impurities.

# 2. Measurement

An analytical technique for the separation and measurement of Pu 239 and Pu240 is described in [NUR 85] (see U 234 chapter).

#### 3. Data and Correlation

In all measurements available, the activities for Pu 239 and Pu 240 are given in a combined form as the total activity of Pu 239 and Pu 240. This is common practice as the spectra of both nuclides cannot be separated in normal alpha spectrometric measurements after radiochemical separation.

It has been found that many TRU nuclides can be correlated to Pu 239+Pu 240 very well. But as it is intended to establish a correlation between TRU nuclides and the easy-to-measure nuclides Cs 137 or Co 60, correlations between Pu 239 and Cs 137 or Co 60 have to be calculated.

Data are available from Sweden, Germany and USA in the form of combined activity of Pu 239 and Pu 240.

The correlations with Cs 137 are of the form:

 $A_{Pu 239+Pu 240} = 3.1 \cdot 10^{-4} \cdot (A_{Cs 137})^{0.81}$  for PWR and

 $A_{Pu 239+Pu 240} = 0.05 \cdot (A_{Cs 137})^{0.62}$  for BWR

and are presented in the first and second diagram.

For the correlations with Co 60 data are available from Germany and Sweden. The correlations are generally better. They have the form:

 $A_{Pu 239+Pu 240} = 2 \cdot 10^{-6} \cdot (A_{Co 60})^{1.1}$  for PWR and

 $A_{Pu 239+Pu 240} = 3 \cdot 10^{-5} \cdot (A_{Co 60})^{0.84}$  for BWR

and are presented in the third and fourth diagram.

Scattering of data is quite large except for the correlation of BWR data with Co 60 which is reflected in the r values and the error ranges.

The country specific analysis in the following table confirms these observations. No country specific peculiarities are observed. For the PWR and BWR correlations with Co 60 it is important to note that the German and Swedish data sets cover different activity regions as was the case for Pu 238 (for PWR: Sweden low activities, Germany higher activities, for BWR vice versa; cf. diagrams). Therefore, the resulting correlations of the combined data sets must be interpreted with caution.



Reactor	Country	a	b	r	S	F	n	A <sub>Pu 239/240</sub> =
PWR	Sweden	$0.83 \pm 0.05$	$-4.5 \pm 0.1$	0.94	0.6	253	35	$3 \cdot 10^{-5} \cdot (A_{Cs \ 137})^{0.83}$
Cs 137	Germany	0.61 ± 0.07	$-2.8 \pm 0.3$	0.75	1.00	81	66	$1.6 \cdot 10^{-3} \cdot (A_{Cs \ 137})^{0.61}$
	USA	$0.66 \pm 0.05$	$-2.8 \pm 0.2$	0.72	1.51	162	150	$1.6 \cdot 10^{-3} \cdot (A_{Cs \ 137})^{0.66}$
	all	0.80 ± 0.03	$-3.5 \pm 0.1$	0.84	1.4	587	251	$3.2 \cdot 10^{-4} \cdot (A_{Cs \ 137})^{0.80}$
BWR	Germany	$0.6 \pm 0.2$	$-3.1 \pm 0.6$	0.62	1.4	17	29	$8 \cdot 10^{-4} \cdot (A_{Cs \ 137})^{0.6}$
Cs 137	USA	$0.62 \pm 0.04$	$-2.1 \pm 0.2$	0.85	0.94	279	112	$8 \cdot 10^{-3} \cdot (A_{Cs \ 137})^{0.62}$
	all	0.62 ± 0.04	-2.3 ± 0.2	0.79	ŢŢŢŢ	228	142	$5 \cdot 10^{-3} \cdot (A_{Cs \ 137})^{0.62}$
PWR	Sweden	$1.00 \pm 0.04$	$-6.4 \pm 0.1$	0.97	0.44	573	38	$4 \cdot 10^{-7} \cdot (A_{Co  60})^{1.00}$
Co 60	Germany	0.64 ± 0.07	$-3.4 \pm 0.3$	0.74	0.99	86	75	$4 \cdot 10^{-4} \cdot (A_{Co60})^{0.64}$
	all	1.09 ± 0.05	-5.7 ± 0.2	0.91	1.18	534	113	$2.10^{-6} \cdot (A_{Co60})^{1.09}$
BWR	Sweden	$0.86 \pm 0.06$	$-4.8 \pm 0.7$	0.95	0.52	184	21	$1.6 \cdot 10^{-5} \cdot (A_{Co 60})^{0.86}$
Co 60	Germany	$0.7 \pm 0.1$	$-4.3 \pm 0.6$	0.73	1.23	34	32	5·10 <sup>-5</sup> ·(A <sub>Co 60</sub> ) <sup>0.7</sup>
	all	0.83±0.04	~4.5 ± 0.3	0.95	1.00	508	<u>53</u>	$3.2 \cdot 10^{-5} \cdot (A_{Co \ 60})^{0.83}$
a = slope, $b = y$ intersection, $r = correlation coefficient$ , $s = standard deviation$ , $F = F$ -test, $n = number of data points$								

## 4. Discussion

The quality of the correlations with Cs 137 is rather poor with values of r around 0.8, but F high enough for statistical significance. It is possible that no generic correlation exists, but only on a plant specific basis. A correlation with Co 60 seems to give better results (r and F slightly higher), but probably has to be carried out also on a plant specific basis as is indicated by the country specific analysis.

Correlation between Pu 239+Pu 240 and Cs 137, PWR



log(Cs 137 [Bq/g])

Pu 239, Pu 240

Correlation between Pu 239+Pu 240 and Cs 137, BWR



2 0 log(Pu 239+Pu 240 [Bq/g]) -2 Pu 239/240, Sweden 3 Pu 239/240, Germany ٩ Regression region of conf. - -- region of conf. -4  $y = 1.085^*x + (-5.691)$  $a = 1.085 \pm 0.047$ b = -5.691 ± 0.185 -6 r = 0.91 F = 534s(x.y) = 1.18 Qx = 631n = 113 -8 -2 -1 0 1 2 3 5 6 7 8 4 log(Co 60 [Bq/g])

#### Correlation between Pu 239+Pu 240 and Co 60, PWR





Correlation between Pu 239+Pu 240 and Co 60, BWR



**Pu 241** 

# 1. Properties and Formation

T <sub>12</sub>		14.3 a	
Decay mode:		$\alpha: \stackrel{241}{_{94}}Pu \xrightarrow{\alpha} \stackrel{237}{_{92}}U$ $\beta^{-}: \stackrel{241}{_{94}}Pu \xrightarrow{\beta^{-}} \stackrel{241}{_{95}}Am$	
Radiation	y(i) [(Bq·s) <sup>-1</sup> ]	E(i) [MeV]	y(i) • Ē(i)
β-	1	5.236E-03	5.24E-03

(y(i), E(i), y(i)  $\cdot$  E(i): see beginning of appendix D for explanation)

Pu 241 is formed from Pu 240 by irradiation with thermal neutrons. Pu 241 belongs to the actinides that are formed by burning uranium fuel. Sources for Pu 241 are the fuel elements and uranium material impurities.

## 2. Measurements

An analytical technique for the separation and measurement of Pu 241 is described in [NUR 85]. For the first steps, see U 234 chapter. The following steps are described as follows:

A fraction from the separated Pu is counted in a liquid scintillation counter. Pu 241 is counted in the tritium window and alpha-emitting Pu is counted in a window above tritium. Pu 236 tracer yield is determined from the ratio of Pu 236 to total alpha/plutonium applied to the liquid scintillation alpha count.

# 3. Data and Correlation

As for Pu 238, a good correlation of Pu 241 with Pu 239+Pu 240 can be assumed.

Data are available for Germany and USA. At first, a tentative correlation analysis with Cs 137 is performed. From the diagrams it can be seen that for PWR and BWR the data scatter widely. The correlations are of the form:

 $A_{P_{u} 241} = 0.2 \cdot (A_{Cs 137})^{0.61}$  for PWR and

 $A_{Pu \ 241} = 0.4 \cdot (A_{Cs \ 137})^{0.64}$  for BWR

and are shown in the first and second diagram.

Pu 241 - 1

The quality of these correlations is rather poor with values of r between 0.75 and 0.8 and F about 250 indicating statistical significance. It is to be expected that no generic correlation exists but has to be carried out on a plant specific basis.

In addition, the correlation of Pu 241 with Pu 239+Pu 240 is investigated. For this correlation, only data from Germany are available. As was expected, the correlation between Pu 241 and Pu 239+Pu 240 is of very good quality (r near 1, F comparatively high). The correlation is of the form

 $A_{Pu\ 241} = 120 \cdot (A_{Pu\ 239+Pu\ 240})^{0.83}$ 

and is shown in the third diagram.

These observations are confirmed by the country specific analysis in the following table, which shows quite good agreement between PWR data from Germany and USA (similar exponent, identical pre-factor, similar quality of correlation). BWR data are available nearly exclusively from USA and show similar results as for PWR.

Reactor	Country	<b>a</b> 21/4 1. A 21/4 1.	<sup>1</sup> ∂∦, b ∖∖	*( <b>r</b> - )	S.	F	` <b>n</b> < <sup>1</sup>	$A_{Pu \ 241} =$	
PWR	Germany	$0.7 \pm 0.1$	$-0.7 \pm 0.4$	0.77	1.05	34	25	$0.2 \cdot (A_{Cs \ 137})^{0.7}$	
Cs 137	USA	$0.60 \pm 0.04$	$-0.7 \pm 0.1$	0.75	1.1	236	184	$0.2 \cdot (A_{Cs137})^{0.60}$	
	all	0.61 ± 0.04	$-0.7 \pm 0.1$	0.75	1.1	268	209	0.2·(A <sub>Cs 137</sub> ) <sup>0.61</sup>	
BWR	USA	$0.64 \pm 0.04$	$-0.4 \pm 0.2$	0.82	0.90	243	124	$0.4 \cdot (A_{Cs137})^{0.64}$	
Cs 137	all	0.64 ± 0.04	$-0.4 \pm 0.2$	0.82	0.90	244	125	$0.4 \cdot (A_{Cs \ 137})^{0.64}$	
all, Pu 239/240	Germany	0.83 ± 0.04	2.07 ± 0.07	0.97	0.36	507	31	0.2·(A <sub>Pu 239/240</sub> ) <sup>0.83</sup>	
a = slope.	a = slope, $b = y$ intersection, $r = correlation coefficient$ , $s = standard deviation$ , $F = F$ -test, $n = number of data points$								

#### 4. Discussion

The quality of the correlation between Pu 241 and Pu 239 + Pu 240 is quite good, which is in accordance with the results from Pu 238. For this correlation, no distinction need to be made between PWR and BWR. The exponent of 0.83, however, in the correlation between Pu 241 and Pu 239/240 cannot be easily explained. An exponent of 1 as for Pu 238 would have been expected. Additional data should be evaluated before a final correlation equation can be established.

Any correlation between Pu 241 and Cs 137 or Co 60 should not be calculated directly but via the correlation between Fu 241 and Pu 239/240, which is of very good quality, and the correlation between Pu 239+Pu 240 and Cs 137 (see chapter on Pu 239+Pu 240 above).

Correlation between Pu 241 and Cs 137, PWR





Pu 241

Correlation between Pu 241 and Cs 137, BWR



Correlation between Pu 241 and Pu 239+Pu 240





Pu 241



# 1. Properties and Formation

1	432,2 a							
Decay mode:	$\alpha: \stackrel{241}{_{95}}Am \xrightarrow{\alpha} \stackrel{237}{_{93}}Np$							
Radiation	y(i) [(Bq:s) <sup>-1</sup> ]	E(i) [MeV]	y(i) · E(i)					
α	1.28E-01	5.443	6.97E-01					
α	8.52E-01	5.486	4.67E+00					
γ	2.40E-02	2.634E-02	6.32E-04					
γ	3.57E-01	5.954E-02	2.13E-02					

 $(y(i), E(i), y(i) \cdot E(i))$ : see beginning of appendix D for explanation)

Am 241 is formed from Pu 240 by irradiation with thermal neutrons. Am 241 is contained in irradiated uranium. Sources are fuel elements and uranium material impurities.

# 2. Measurement

An analytical technique for the separation and measurement of Am 241 is described in [NUR 85]. See the chapter on U 234 for details.

# 3. Data and Correlation

As for Pu 238, a good correlation with Pu 239+Pu 240 can be assumed.

From the diagrams it can be seen that for PWR and BWR the data scatter widely, especially for BWR. The correlations are of the form

 $A_{Am 241} = 5 \cdot 10^{-5} \cdot (A_{Cs 137})^{0.95}$  for PWR and

 $A_{Am 241} = 3 \cdot 10^{-2} \cdot (A_{Cs 137})^{0.3}$  for BWR

and are shown in the first and second diagram. For PWR, the correlation is of medium quality with r=0.92 and F indicating statistical significance. For BWR, however, no correlation exists (r=0.27, F very low).

In addition, the correlation of Am 241 with Pu 239+Pu 240 is investigated. For this correlation, data from Sweden and Germany are available. As was also found for Pu, the correlation between

Am 241 and Pu 239+Pu 240 is of very good quality (r near 1, F very high) and apparently need not be distinguished for PWR and BWR. The correlation is of the form

 $A_{Am\ 241} = 0.9 \cdot (A_{Pu\ 239+Pu\ 240})^{1.01}$ 

and is shown in the third diagram.

The country specific analysis in the following table support the results discussed above.

Reactor	Country	a	b	ŕ.	S	F	'n	$A_{Am241} =$
PWR	Sweden	$0.84 \pm 0.06$	$-5.2 \pm 0.1$	0.94	0.70	206	30	$6 \cdot 10^{-6} \cdot (A_{Cs \ 137})^{0.84}$
Cs 137	Germany	$0.55 \pm 0.06$	$-2.4 \pm 0.3$	0.74	0.94	77	65	$4 \cdot 10^{-3} \cdot (A_{Cs \ 137})^{0.55}$
na Nasimpo	all	0.95 ± 0.04	$-4.3 \pm 0.2$	0.92	1.21	514	95 ,	$5 \cdot 10^{-5} \cdot (A_{Cs \ 137})^{0.95}$
BWR	Germany	$0.2 \pm 0.2$	$-1.4 \pm 0.7$	0.25	1.1	2	29	$4 \cdot 10^{-2} \cdot (A_{Cs \ 137})^{0.2}$
	all	$0.3 \pm 0.2$	$-1.6 \pm 0.7$	0.27	1.2	2.2	31	$3 \cdot 10^{-2} \cdot (A_{Cs \ 137})^{0.3}$
all	Sweden	$1.02 \pm 0.02$	$-0.5 \pm 0.1$	0.99	0.77	2642	45	$0.3 \cdot (A_{Pu \ 239/240})^{1.02}$
Pu 239/	Germany	0.79 ± 0.05	$0.02 \pm 0.07$	0.86	0.65	272	97	$1 \cdot (A_{Pu} _{239/240})^{0.79}$
Pu 240	all	1.01 ± 0.02	$-0.07 \pm 0.07$	0.98	0.79	2907	142	$0.9 \cdot (A_{Pu \ 239/240})^{1.01}$
a = slope, $b = y$ intersection, $r = correlation coefficient$ , $s = standard deviation$ , $F = F$ -test, $n = number of data points$								

#### 4. Discussion

A good correlation between Am 241 and Pu 239+Pu 240 has also been found in [NUR 85]. There it is estimated that this correlation should provide useful generic scaling factors. It is also pointed out that the Am activity is less abundant than the Pu activities and that any larger variance in the scaling factor is therefore less important than it would be for Pu. The scaling factor found in [NUR 85] was 0.62 for BWR and 0.48 for PWR which is less than what was calculated in the present study (0.9). All the same the good quality of the correlation for BWR and PWR that was found in this study supports the view expressed in [NUR 85].

If the Am 241 contents is to be assessed from the key nuclides Co 60 and Cs 137, it can therefore be recommended to use the correlation between Am 241 and Pu 239+Pu 240 and the correlation between Pu 239+Pu 240 and Cs 137 (see chapter on Pu 239+Pu 240).

Correlation between Am 241 and Cs 137, PWR



BS



#### Correlation between Am 241 and Cs 137, BWR

log(Cs 137 [Bq/g])

Am 241

Correlation between Am 241 and Pu 239+Pu 240







# Brenk Systemplanung

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