Nuclide Vector for Decommissioning and Release Measurements in Germany

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1. Introduction

One of the key activities before and during decommissioning is the determination of the radiological inventory of the nuclear power plant (NPP). The radiological characterization of the facility is crucial for determining adequate pathways for the release of material from the regulatory control and for the radiation protection of the workers during the decommissioning process. The determination of the radionuclides is the basis for decontamination measures that need to be taken. Germany and many other countries (Korea, US, UK, etc.) use the concept of scaling factors as a convenient way to derive the full radiological profile of nuclear waste [1].

2. German Law for Release of Radioactive Material

In Germany the release of radioactive material is regulated in the radiation protection ordinance (StrlSchV Strahlenschutzverordnung). Attachment 4 table 1 lists the release values for the different nuclides and types of releases (free release and specific release) according to the 10 μSv-concept. If the material contains more than one radionuclide the sum of the ratio of the specific activity (C) and the release value must be smaller than one [5].

$$\sum_i \frac{C_i}{R_i} \leq 1 \qquad (1)$$

with C_i= specific activity of the nuclide i, R_i= relevant release value according to the Radiation Protection Ordinance attachment 4, table 1.

3. Determining a Nuclide Vector

The difficulty for the radiological characterization is that multiple measurement techniques are necessary to determine the exact fractions of alpha-, beta and gamma emitting nuclides. As a result a facility-wide sampling program with a focus on neuralgic points has to be established.

In this section the German method of determining the radiological inventory in NPPs is described and the relevant terms are defined.

3.1 Definition of the Nuclide vector

In Germany the principle of scaling factor is used for release of material from the NPP. The term "nuclide vector" is used commonly and describes the composition of the expected radionuclides in relation to each other [2]. The nuclide vector relates the difficult to measure nuclides (DTM) and easy to measure nuclides (ETM). The scaling factor is a dimensionless factor for the different DTMs which allows to determine the total activity by measuring the activity of the respective ETM [3].

$$A = A_{ETM} \times f_{ETM} \quad (2)$$

with A= total activity in the material, A_{ETM}= activity of the ETM (key nuclide) and f_{ETM}= scaling factor.

The Nuclide vector gives the relative proportions of the activity of the individual nuclides in relation to the total activity.

$$v_i = \frac{a_i}{\sum_i a_i} \qquad (3)$$

with a_i= specific activity of nuclide i and v_i= relative proportion of the nuclide i in the nuclide vector.

Not all radionuclides have the same biological effect. Radionuclides with same activity in Bq differ significantly in their radiological importance which is considered in differing release values [4]. Therefore, another quantity is defined, the weighed and normalized nuclide vector. This quantifies the radiological significance of the different nuclides for the release of waste material from the decommissioning process.

$$\frac{\frac{v_i}{R_i}}{\sum_i \frac{v_i}{R_i}} = w_i \quad (4)$$

with v_i= relative proportion of the nuclide i in the nuclide vector, R_i= relevant release value according to the Radiation Protection Ordinance attachment 4, table 1 [5] and w_i= relative proportion of the nuclide i in the weighed and normalized nuclide vector.

Using the definitions above a nuclide vector for a certain system/room can be established. Table 1 shows the impact of weighing for an exemplary nuclide vector. Notice that Fe-55 is the nuclide with the highest activity; however, it only contributes 0.001% to the weighed nuclide vector (i.e. the risk) whereas, Co-60 and Cs-137 are the main contributors. Also, Ni-63 which is a low energy beta emitter doesn't contribute significantly [4].

Table I: Exemplary (shortened) nuclide vector

Nuclide	v_i in % [4]*	R_i ** [Bq/g]	w_i in %*
Fe-55	42.270801	1000	0.0089487
Co-60	28.851499	0.1	61.0781296
Cs-137	16.774127	0.1	35.5105398
Ni-63	9.39511	100	0.0198859

Eu-152	1.207737	0.1	2.5567586
Ni-59	0.503224	100	0.0010653
Sr-90/Y-90	0.335483	1	0.0710212
Cs-134	0.241547	0.1	0.5113509
Pu-241	0.228128	1	0.0482943
H-3	0.100645	1000	0.0000213
Ag-108m	0.064413	0.1	0.1363612
Eu-154	0.020129	0.1	0.0426127
Am-241	0.003825	0.1	0.0080975

*the values are multiplied with 100 to get %
** Data from StrlSchV Table attachment 4 table 1 [5]

3.2 Determination of the Nuclide vector

In principle determining the nuclide vector for each NPP will follow these basic steps:

1. Determine plant history and "dividing into areas"
2. Selection of the nuclides to be analyzed
3. Creation of a sampling program
4. Sampling
5. Sample preparation
6. Analyzing the samples
7. Calculating the nuclide vector
8. Updating the nuclide vector

The nuclide vector is only representative if the process which lead to the vector (i.e. probe plan, probe taking, probe measurement, etc.) is reproducible and verifiable. Early mistakes in the planning (sampling strategy) and sampling stage (engineer who takes the samples and number of samples taken) have more severe consequences than later mistakes (analysis and calculations) [2].

3.2.1 Plant History and Division into Areas

The first step for determining the nuclide vector is to gather the entire preexisting knowledge of the plant and the plant condition. The activated areas of the plant and the propagation paths (e.g. cooling water circuit, waste water, steam, dust, etc.) should be determined. During this first step it is important to evaluate singular events (e.g. exchange/removal of components, leakages, accidents, etc.) for this it is essential to include the workers, especially experienced workers (even retired workers), who know the early plant history very well.

As a second step the plant will be divided into areas which are expected to have a homogenous nuclide vector. Criteria for "areas" are physical, chemical, material and procedural criteria:

- Buildings/rooms,
- Technical systems,
- Change in material composition,
- Change in surface material
- Etc.

The level of detail for this analysis should be chosen carefully. The ratio of effort and benefit must be considered. A step-by-step approach with increasing level of detail is recommended.

3.2.2 Selection of the Nuclides

Attachment 4, table 1 of the radiation protection ordinance [5] lists the release values for approximately 800 nuclides; however, for the determination of the nuclide vector only the "few" relevant ones are selected. The selection criteria depend on technical data, activation products of the materials used, results from the emission surveillance (aerosols and waste water) and operational history. Results from waste management (i.e. calculation of spent fuel inventory, analysis of evaporator concentrate) are also suitable for the construction of the vector.

The selection of nuclides is important to limit the effort for the analysis, especially the radiochemical analysis. Table 1 of [6] of gives an overview of the relevant nuclides for nuclear power plants.

3.2.3 Sampling Program and Sampling

German operators included the sampling program in the operation manual. It is important to have a clear sampling plan which prevents any subjective ad hoc decisions by the engineer taking the samples. The sampling program/plan includes the following:

- Work instructions/procedure
- Place of sampling
- Time of sampling
- Sampling Engineer
- Sampling method
- Sampling equipment
- Sample preparation
- Sample container
- Sampling protocol
- Measures for QA

For the samples, material samples are preferred. When determining the sampling method, the potential penetration depth of the radionuclides must be considered. Wipe tests have only limited relevance when determining the nuclide vector, as those tests consider only removable contamination. In order to choose the right sampling method, the material and types of radionuclides must be considered to avoid loss of volatile radionuclides, cross contamination and mixing of surface material with base material.

The number of samples necessarily depends on several factors. The number of samples per area must be chosen in such a way to get a 95% confidence level (for statistical evaluation of the nuclide vector 16 to 20 samples seem to be optimal) [2].

3.2.3 Sample Preparation and Analysis

For the sampling preparation it needs to be ensured that the nuclide vector is not changed by e.g. loss of volatile radionuclides, cross contamination etc. Clear liquids are easy to handle, for opaque liquids it may be necessary to separate solid and liquid parts for the analysis. Sludges are dried for analysis and treated like

solid samples. Solid samples are crushed, homogenized and separated and then analyzed.

For the sample analysis it is recommended to analyze the sample with the lowest specific activity first and then in the order of increasing activity, in order to prevent contamination. All samples should be analyzed by gamma spectrometry which is less work than other methods. Based on those measurements approximately 10% of the samples are analyzed radiochemically to determine the alpha and beta radiation [2].

3.2.3 Calculating the Nuclide vector

All data from the measurements should be validated before using them for calculation of the vector (i.e. completeness, plausibility, reliability of the data, etc.). For the calculation, the specific activities of all nuclides in one sample are summed up and then the share of each nuclide is determined.

There are three different approaches to calculate a nuclide vector:

- Abdeckender (covering) Nuclide vector (conservative but not representative)
- Nuclide vector based on statistics (conservative and representative)
- Nuclide vector based on mean value formation (representative but not conservative)

Covering nuclide vectors:

The covering nuclide vector is determined by calculating the most unfavorable relation of the nuclides. Unfavorable means on the one hand to maximize the relation of the specific activity and the release values in the summation in equation (1) and to maximize the scaling factors. From all the analysis results of the samples the maximum measured value for each DTM nuclide is taken and then the remaining activity is assigned to the key nuclide. This leads to significant overestimation of the non-measurable nuclides. Therefore, the covering nuclide vector is conservative, but it is not representative (doesn't represent the correct relation between the key nuclide and the DTM nuclides). There are several ways to reduce the overestimation of DTM nuclides in the covering nuclide vector, one method is to group nuclides and assign one key nuclide for each group. For example [2]:

- Activation products (key nuclide Co-60)
- Fission products (key nuclide Cs-137+)
- Transuranic elements (key-nuclide Am-241)

The covering nuclide vector is straight forward to calculate and nuclide vectors which are valid for a larger area can be determined. The disadvantage of covering vectors is that they are overestimating the activity of the material and therefore release limits can be easily exceeded, particularly for alpha emitters.

Statistic nuclide vector:

For every area of the plant where a homogenous contamination is expected samples are taken (for statistical relevance more than 16 samples are necessary per area, depending on area size and included systems). It is then statistically analyzed if the data is distributed according to a log or normal distribution.

This approach has been used for the NPP in Stade and the NPP in Würgassen. As a first step the nuclide vector of every sample belonging to one predefined "area" is determined. As a next step for every radionuclide the mean and the standard deviation is determined.

The contribution of each radionuclide is now variated within one-sigma of the mean value

$$(\bar{x} - \sigma) \leq x \leq (\bar{x} + \sigma) \quad (5)$$

with $\bar{x}$ the mean value, σ the standard deviation and x the value. The variation is done in a way to maximize the following three terms [2]:

- Maximize the sum of equation (1)

$$\sum_i \frac{C_i}{R_i} = maximum \quad (6)$$

with C_i= specific activity of nuclide i and R_i= relevant release value according to the Radiation Protection Ordinance attachment 4, table 1 [5].

- Maximize the proportion (as defined in equation 3) of DTM nuclides (alpha and beta emitters without well measurable gamma lines) in the nuclide vector.

$$\sum_i v_{\propto+\beta,i} = maximum \ (7)$$

with $v_{\propto+\beta,i}$= relative proportion of the alpha and beta radiation nuclide i in the nuclide vector.

- Maximizing the sum related to the area-related activity according to the Radiation Protection Ordinance attachment 4 [5]

$$\sum_i \frac{C_i}{O_i} = maximum \ (8)$$

with C_i= specific activity of nuclide i and O_i= relevant area-related release value according to the Radiation Protection Ordinance attachment 4, table 1 [5].

All three terms cannot be maximized at the same time therefore the goal is to maximize the sum value. This will then determine the degree of conservativity of the nuclide vector for each of those three areas.

The advantage of this type of nuclide vector is that it is representative and conservative. The disadvantage of this method is that a relatively large number of samples is required, and the calculations are more extensive than in the case of the covering nuclide vector [2]. The nuclide

vectors are only valid for medium sized areas or certain systems.

Mean value nuclide vector:

The nuclide vector can be determined by simply taking the mean of the analysis results of the samples of one "area". For this method it is necessary to have a sufficient number of samples and a homogenously spread contamination in order to ensure statistical relevance of the results.

Using the mean value method will give representative but not conservative nuclide vectors. For samples some portion of the values will be higher than the ones predicted by the nuclide vector.

The advantage of this method is that the calculation is straight forward and nuclide vector which are valid for larger "areas" can be determined. The disadvantage is that the vectors are not conservative, and corrections have to me made for the release of material [2]. The nuclide vectors are only valid for small sized areas or single systems, so there will be a large number of nuclide vectors for a whole plant.

3.2.3 Updating Nuclide vector

The nuclide vectors always need a reference date to check their validity and adjust them according to the decay scheme of relevant radionuclides. Updating the nuclide vector also needs to be considered, e.g. after a full system decontamination or if there are discrepancies in the release measurement. For German NPPs nuclide vectors with a large contribution from Co-60 (half-live 5.3a) are updated according to the decay scheme every two years [2].

4. Conclusions

Determining the nuclide vector is challenging but essential for the decommissioning of nuclear power plants and particularly for the release of radioactive material from the regulatory control. The key is to have qualified personnel to establish a sampling plan and to take the samples. Mistakes made in this first steps cannot be compensated for later.

It is important to divide the plant into "areas" according to expected nuclide composition, system borders, and others. The analysis of the samples is done in accredited laboratories (e.g. Framatome) outside the plants or in laboratories of the operator (e.g. for gamma spectrometry).

There are three basic approaches to calculate the nuclide vector (covering, statistic, mean-value), each method has their advantages and disadvantages which method is selected depends on the operator, the condition of the plant and other factors.

The nuclide vectors should always have a valid date and be updated if necessary (after certain time, full system decontamination, etc.).

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