Opportunities and limits of Decay storage based on Dismantled Materials in Nuclear Power Plants - 17650

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ABSTRACT

We investigate the potential of decay storage of low and intermediate radioactive waste for release procedures. We present the Safetec software tool HL3 and its application to data from the German nuclear power plant at the site in Stade.

In general, during the dismantling process of nuclear power plants, waste of slightly contaminated and/or activated systems is produced in large amounts. These are in particular demolition rubble from concrete structures and ingots from the melting process of metals. If, at the time of disposal, the nuclear inventory is near or slightly above federal clearance limits, the material cannot be released as conventional waste.

However, due to the decline of the nuclear inventory over time, a significant amount of that waste could qualify for the release procedure in the future. Although this concept of decay storage is well known, specific predictions of the mass flow to be released is non-trivial. The mass flow over time depends on the composition of known and assumed nuclides (nuclide vector), as well as on the underlying threshold values for release.

To handle the waste stream as a function of time, the nuclide composition and specific threshold values, Safetec developed a bespoke software tool named HL3. Its primary function is to visualize and to test/analyze various nuclide vectors and its decompositions over time. For the application within German law, threshold values according to the different disposal routes can be considered. Using the batch interface in the software tool HL3, large amounts of data can be automatically analyzed in a traceable and quick way.

To test both, the tool and the benefit for the concept of decay storage, we have applied our code HL3 to material from the dismantling of the pressurized-water reactor at the site in Stade, Germany.

We set the focus on concrete debris and metal ingots, which in principle are best suited for decay storage over years. The assignment and the packaging of that waste from Stade were based on application documents for decommissioning of the Year 2000 and according to the state of art conditioning technique at that time. Based on the data of that waste and the appropriate disposal routes, we calculate and discuss the amount of waste releasable after decay storage.

As for the situation in Germany, we find that only a negligible amount would be releasable, mainly due to other reasons. These are the deterioration of detectability of radionuclides with time, and other regulatory conditions. However, on an international scale, a different assessment can be drawn. Our results thus motivate to reconsider the option of interim decay storage to reduce radioactive waste in large amounts.

INTRODUCTION: NUCLIDE VECTORS AND THRESHOLD VALUES

The dismantling of nuclear power plants represents a challenging task. In particular, the large amounts of low-level radioactive waste have to be disposed according to different disposal routes. In principle, these are either disposal in final repositories as nuclear waste, or the unrestricted release as conventional waste. While various other issues and parameters must be considered before the waste can be classified, the most important aspect is the radionuclide inventory of the waste, primarily a function of the waste generating process.

To determine the nuclide composition, either a representative and realistic nuclide vector (NV) is used, or at least a conservative nuclide vector, which covers all uncertainties in a sufficient way [1]. The nuclide vector comprises of the nuclide ratios to at least one (or more) easy to measure nuclide(s). The typical reference nuclides used are e.g. Co-60 and Cs-137, for activation and/or contamination paths respectively. So once the activity of the reference nuclide(s) is measured for a given waste entity, the other nuclide activities follow directly from its ratios in the corresponding nuclide vector. In this manner, a high mass throughput can be achieved in a comprehensive and technically rather simple way.

As for the situation in Germany, one prerequisite for release as conventional waste is that the federal limits for the non-background activities must not be exceeded. In the presence of several nuclides n, the sum S of the nuclide ratios must be equal to or smaller than 1,

$$S = \sum_{i=1}^{n} \frac{a_i}{F_i} \leq 1, \ (i=1, \ \dots \ ,n), \qquad \text{(Eq. 1)}$$

where a_i is the specific activity of the i-th nuclide (here in Bq/g), and F_i is the federal limit for the corresponding i-th nuclide.

As for the situation in Germany, the threshold values (limits) are in addition defined according to different disposal routes. Here we will focus on the disposal of demolition debris for landfill (waste disposal site) up to 100 Mg/a and 1000 Mg/a, respectively, as well as its unrestricted release. The German federal limits are listed in the “Strahlenschutzverordnung” (StrlSchV) [2].

If a waste package exceeds the sum S >1, see (Eq. 1), it cannot be released and has to be conditioned for a final repository. This in general will be the more cost extensive path. Above all, it should be noted that in Germany in particular, and in other countries in general, the amount of radioactive waste that can be disposed of will be limited to space and radiological capacities of available repositories.

THE CONCEPT OF DECAY STORAGE AND OUR ANALYSIS TOOL HL3

Since the waste, which is subject to final storage, has to be placed in intermediate on-site facilities in any way, in this work we will analyze and evaluate the concept of “decay storage”: The radionuclide inventory will decline over time, eventually allowing a release as conventional waste sometime in the future. Especially low-level waste with activities near or slightly above the release limits is a good candidate for this concept. Such waste often constitutes of rather short and intermediate-lived radioactive nuclides, like Co-60 (5.3 years half-life) and Cs-137 (30.2 years half-life), respectively.

There are many other technical difficulties to achieve successful decay storage, in particular constraints from measurements and regulatory framework. However, in this article we will only focus on the influence of the nuclide vectors and the underlying/corresponding threshold values, to determine the time when the criteria S≤1, see (Eq. 1), for low-active waste is met.

For the analysis of large amounts of waste packs, and the influence of various nuclide vectors, we have developed the computer tool "HL3" [3]. We show its application first for a general observation of the concept before investigating the most promising candidates for decay storage, which are demolition rubble and metal ingots.

Input quantities to our analysis tool HL3 are the nuclide vector, its reference time, the activity of the reference nuclide with its corresponding date of measurement, the mass of the given waste, as well as the intended disposal route. (The chosen disposal route will determine the corresponding threshold values to be selected for the analysis.)

Solving the Bateman equations both for nuclides which decline and those which will build-up allows any of the nuclides in the nuclide vector to be determined at a given time t. A specific solution of the Bateman equation for the decay-chain $X_1 \rightarrow X_2 \rightarrow X_3$ yields the activity of the parent nuclides

$$A(t) = A_0 e^{-\lambda t}, \qquad \text{(Eq. 2)}$$

and the activity of the daughter nuclides

$$A_2(t) = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1^0 \left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + A_2^0 e^{-\lambda_2 t}, \qquad \text{(Eq. 3)}$$

where the λ_is are the decay constants. In the code "HL3" we have included the important build up of the alpha emitter Am-241 due to the decay of the beta-emitter Pu-241.

For any given waste, the tool correlates the activities to the mass and calculates the year when the condition for the sum S=1 is met, see (Eq. 1). The output returns the activities of the nuclides in the nuclide vector for that time and its ratios in respect to the federal limits.

Clearance after decay storage

As an application of the tool HL3, we first analyze selected data from the low-level active waste in the intermediate storage facility LarA, at the nuclear power plant at the site in Stade, Germany (KKS). Stade was one of the first nuclear power plants which was shut-down in 2003 after the German nuclear power phase-out legislation. After proper conditioning, the most part of the dismantled material for KKS has already been released according to federal standards. The remaining radioactive waste from dismantling which did not meet the clearance criteria was stored in the interim storage facility LarA, waiting for the opening of the repository KONRAD in Germany.

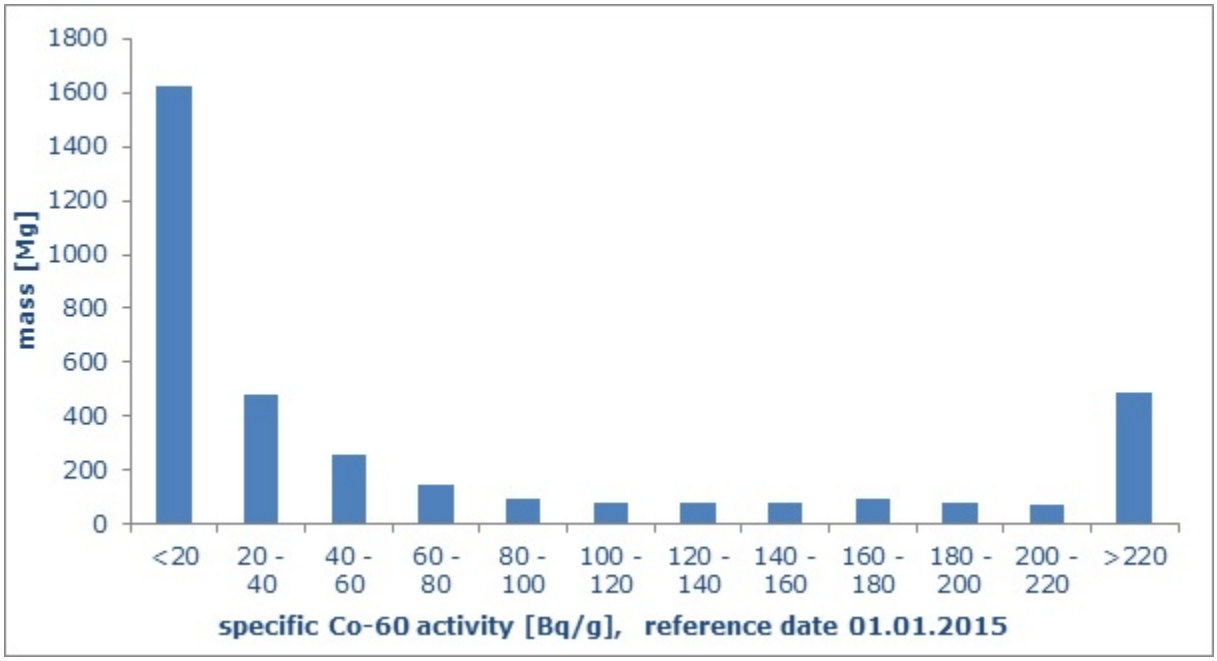

Fig. 1: Activity distribution of the waste in the intermediate storage facility LarA, Stade, Germany

For an overview of the activity inventory in the waste stored in the intermediate storage facility, the distribution of the specific Co-60 activity over the mass is shown in Fig. 1. The data excludes waste of melted metal scrap (to be discussed separately) and intermediate level waste with high dose rate, which is packed and stored in MOSAIK containers.

For the calculation of the time periods for decay storage, we have to fix the nuclide vectors (NV) to define the nuclide decomposition. For illustration, we choose both vectors for a pressurized water reactor (PWR) and for a boiling water reactor (BWR). Note that our chosen values can only give a flavor and orientation for a possible nuclide distribution. In particular, the given nuclide vectors have to be considered conservative for those nuclides which are less accessible, i.e., which are more difficult to measure, and/or show a wider distribution in representative samples. Individual values of the nuclides and also the inclusion of additional nuclides other than those mentioned can be considered for any further or independent analysis.

Nonetheless, we present valuable NVs for PWR and BWR. They are extracted from samples of evaporator bottom. In TABLE I we list the occurring nuclides, their corresponding half-lives $T_{1/2}$, and the ratios in the nuclide vector

$$NV_i = \frac{A_i}{A} \qquad \text{(Eq. 4)}$$

where the activity A_i of the i-th nuclide (in Bq) and the total activity $A = \sum_i A_i$.

In the last two columns of TABLE I we also give the weighted (and normalized) nuclide vector [1]

$$\text{weight. } NV_i = \frac{s_i}{s} \qquad \text{(Eq. 5)}$$

TABLE I: Decline of nuclear inventory and the normalized nuclide vector of a pressurized water reactor (10 years after shut down) in respect of federal limits

nuclides	half-life $T_{1/2}$ [a]	nuclide vector NV 01/01/2015	weight. NV 01/01/2015	weight. NV 01/01/2055
Fe-55	2.7	1.0%	3.9E-03%	1.8E-06%
Co-60	5.3	11%	86%	6.2%
Ni-63	100	80%	0.21%	2.1%
Sr-90	28.5	0.30%	0.39%	2.0%
Ag-108m	127	0.20%	0.78%	8.5%
Sb-125	2.8	0.10%	0.098%	6.6E-05%
Cs-137	30.2	7.0%	11%	59%
Pu-238	87.7	0.020%	0.39%	3.9%
Pu-239/(240)	24000	0.020%	0.39%	5.3%
Pu-241	14.4	0.30%	0.12%	0.23%
Am-241	432.6	0.050%	0.78%	12%
Cm-243/(244)	29.1	0.010%	0.11%	0.59%
	sum:	100%	100%	100%

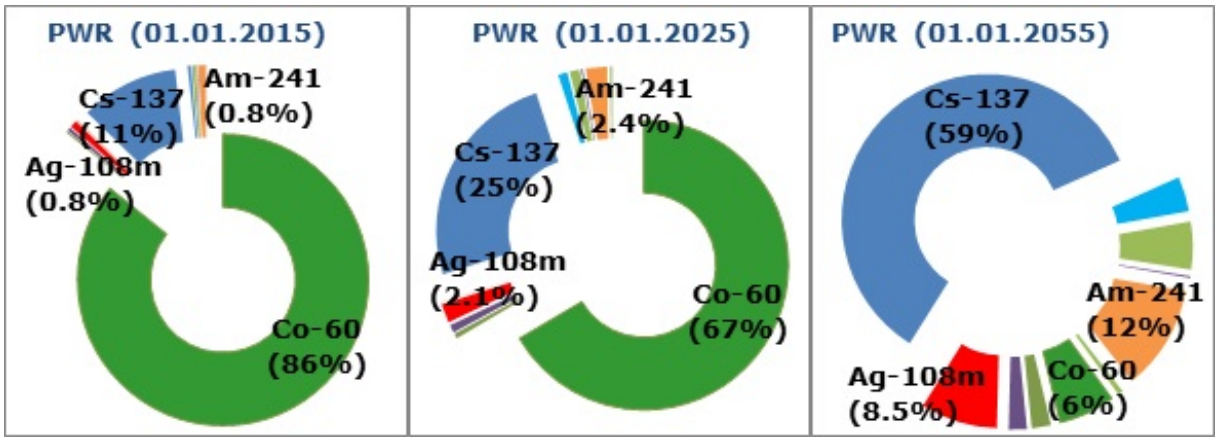

Fig. 2: Weighted and normalized PWR nuclide vector for unrestricted clearance

where the sum values S_i and $S = \sum_i S_i$ as given in (Eq. 1). The weighted and normalized nuclide vector takes into account the federal threshold values, und thus illustrates the importance of the different nuclides for the release procedure. As we can see from TABLE II, depicted graphically in Fig. 2, the importance of the nuclide Co-60 in our vectors will decrease in favor of Cs-137, Sr-60, Ag-108m and alpha emitters, in particular Am-241 due to its long half-life and due to the build up from Pu-241 decay.

The threshold values F_i for clearance procedures are taken from the German radiation protection ordinance (StrlSchV) for unrestrictive clearance (see *table 1 column 5 of annex 3 of the StrlSchV*) [2].

In comparison, our nuclide vector for a boiling water reactor (see TABLE II and Fig. 3) clearly differs in the composition of the radioactive nuclides like Fe-55, Co-

60, and Ni-63. This will strongly influence the composition of the nuclide vector in the year 2055. The proportion of Co-60 in the PWR nuclide vector is enhanced in comparison to the PWR nuclide vector, which in turn has a greater share of Am-141 and Ag-108m.

TABLE II: Decline of nuclear inventory and the normalized nuclide vector of a boiling water reactor (about 3 years after shut down) in respect of federal limits

nuclides	half-life $T_{1/2}$ [a]	nuclide vector NV 01/01/2015	weight. NV 01/01/2015	weight. NV 01/01/2055
Fe-55	2.7	36%	0.039%	6.9E-05%
Co-60	5.3	44%	97%	26%
Ni-63	100	12%	0.008%	0.32%
Sr-90	28.5	0.42%	0.15%	2.9%
Sb-125	2.8	0.069%	0.019%	4.8E-05%
Cs-134	2.1	0.087%	0.094%	8.8E-06%
Cs-137	30.2	7.0%	3.0%	62%
Pu-238	87.7	0.0023%	0.012%	0.46%
Pu-239/(240)	24000	0.014%	0.077%	3.9%
Pu-241	14.4	0.37%	0.041%	0.30%
Am-241	432.6	0.0081%	0.035%	3.9%
Cm-243/(244)	29.1	0.0021%	0.0065%	0.13%
	sum:	100%	100%	100%

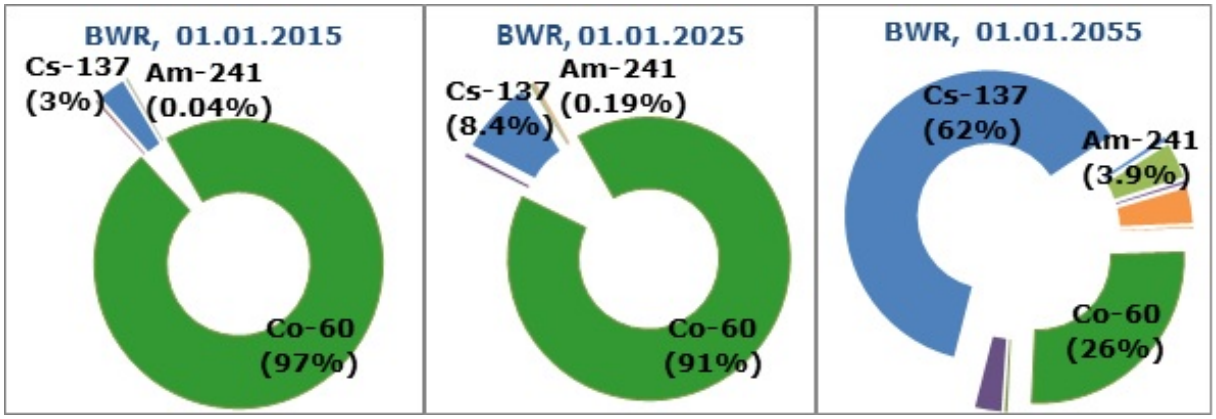

Fig. 3: Weighted and normalized BWR nuclide vector for unrestricted clearance

To analyze the implications of the two nuclide vectors on the concept of storage decay, both were applied to the data shown in Fig. 1. Note, that the nuclide vector might not be generally applicable to that waste. Second, the Co-60 values shown in Fig. 1 are obtained under the assumption that the waste will be disposed of in a final repository. Clearly, these values thus cannot be extrapolated easily to valid activities for clearance. However, we think that still our analysis will be an instructive illustration for the theoretical potential and influence of different compositions of the nuclide vector to the concept of decay storage.

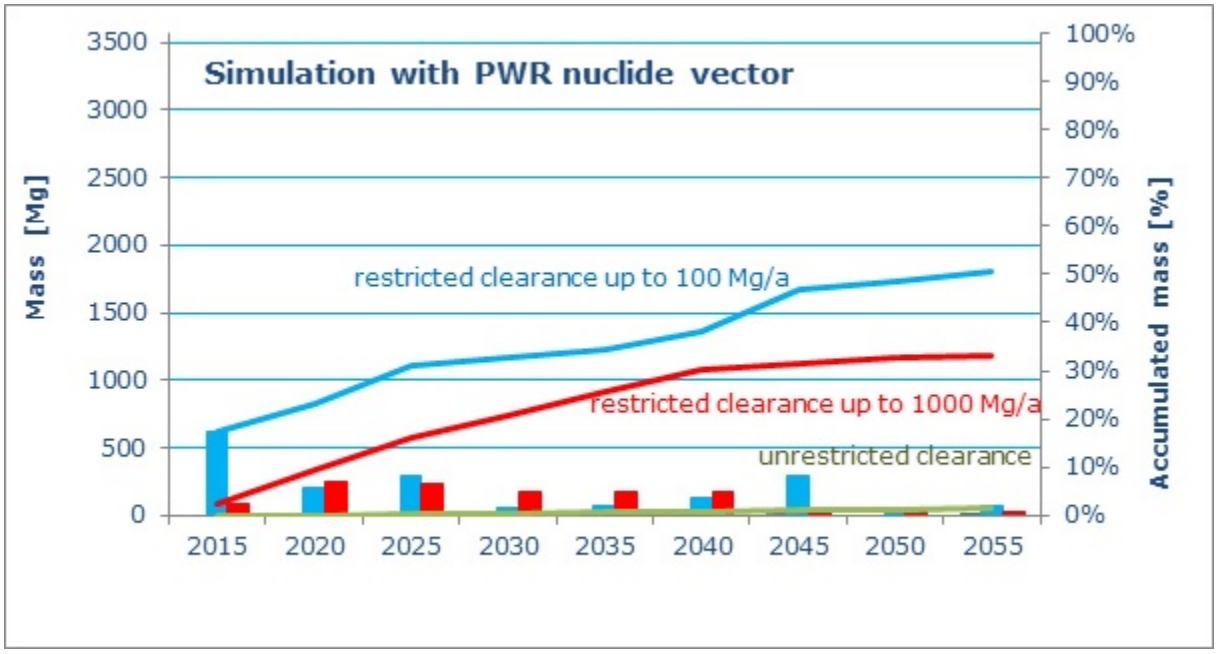

Fig. 4: Clearance after decay storage for the PWR nuclide vector

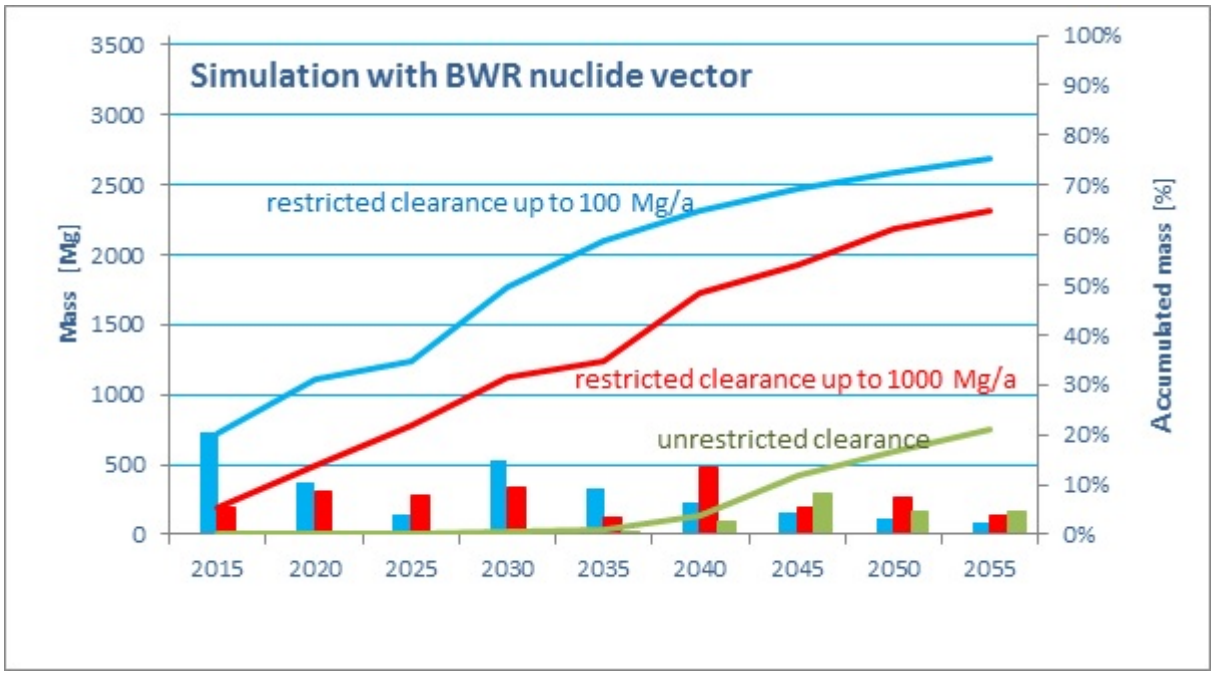

Fig. 5: Clearance after decay storage for the BWR nuclide vector

In Fig. 4 and Fig. 5 we show the mass flow and the cumulated mass over time that could be subject for clearance after the given years of decay storage using the two nuclide vectors for PWR and BWR, respectively. Having applied the specific German clearance levels for deposits (restricted clearance: red and blue; unrestricted clearance: green), a reasonable fraction will be good for clearance. Especially the BWR nuclide vector allows a greater percentage to be cleared. A major factor contributing to this is the ratio of Co-60/Cs -137. The more the ratio is balanced towards Co-60, the more effective decay storage will become due to its rather short half-life. A major key factor for successful decay storage will thus be a good handle to determine the Cs-137 activities in the waste.

Another factor is the proportion of Am-141, which has a rather strict federal limit. However, one should take into account, that beside the criteria fulfilling the clearance levels, other conditions must be met. These criteria are discussed below.

After this rather broad introduction into the topic of decay storage, we now turn to the discussion of waste from construction debris and metal ingots from the process of melting. Due to the inherent properties of concrete and metal, they represent the most promising candidates for a successful decay storage over longer time periods. Moreover, if the melting process is steered effectively, there essentially remains only Co-60 in the ingots, since other nuclides, in particular Cs-137 and the Alpha emitters, are found in the ashes and/or slag, which can be separated.

Demolition debris

During the dismantling process the demolition waste was stored in Big Bags in Stade. Now that waste (in the intermediate storage) has been used to fill up the cavities in the containers with waste-components, which are assigned for the final repository. Other than before, we use a very specific nuclide vector, the so-called VKTA nuclide vector [4]. It was specifically employed for the clearance of contaminated concrete structures/buildings in Stade. We thus correlate it to the debris from concrete structures and decontamination works.

TABLE III: VKTA nuclide vector for contaminated construction waste [4]

nuclides	half-life $T_{1/2}$ [a]	nuclide vector NV 01/01/2010	weight. NV 01/01/2010	weight. NV 01/01/2055
H-3	12.3	28.90%	0.25%	0.12%
C-14	5700	0.09%	5E-03%	0.03%
Mn-54	0.9	0.01%	0.01%	9E-18%
Fe-55	2.7	2.46%	0.01%	4E-7%
Co-57	0.7	1E-03%	2E-04%	9E-22%
Co-60	5.3	10.33%	58.9%	1.02%
Ni-63	100	26.59%	0.05%	0.21%
Zn-65	0.7	2E-04%	3E-04%	8E-24%
Sr-90	28.5	1.70%	1.45%	3.04%
Nb-94	20000	0.02%	0.10%	0.61%
Ag-108m	127	0.05%	0.26%	1.26%
Ag-110m	0.7	2E-03%	0.02%	1E-21%
Sb-125	2.8	0.15%	0.15%	1E-05%
Cs-134	2.1	4E-06%	2E-03%	5E-09%
Cs-137	30.2	28.57%	36.7%	81.7%
Pu-238	87.7	0.04%	0.26%	1.15%
Pu-239/(240)	24000	0.06%	0.37%	2.28%
Pu-241	14.4	0.92%	0.23%	0.17%
Am-241	432.6	0.11%	1.16%	8.40%
Cm-242	0.4	1E-05%	7E-08%	2E-35%

Cm-243 / (244)	29.1	0.01%	0.06%	0.06%

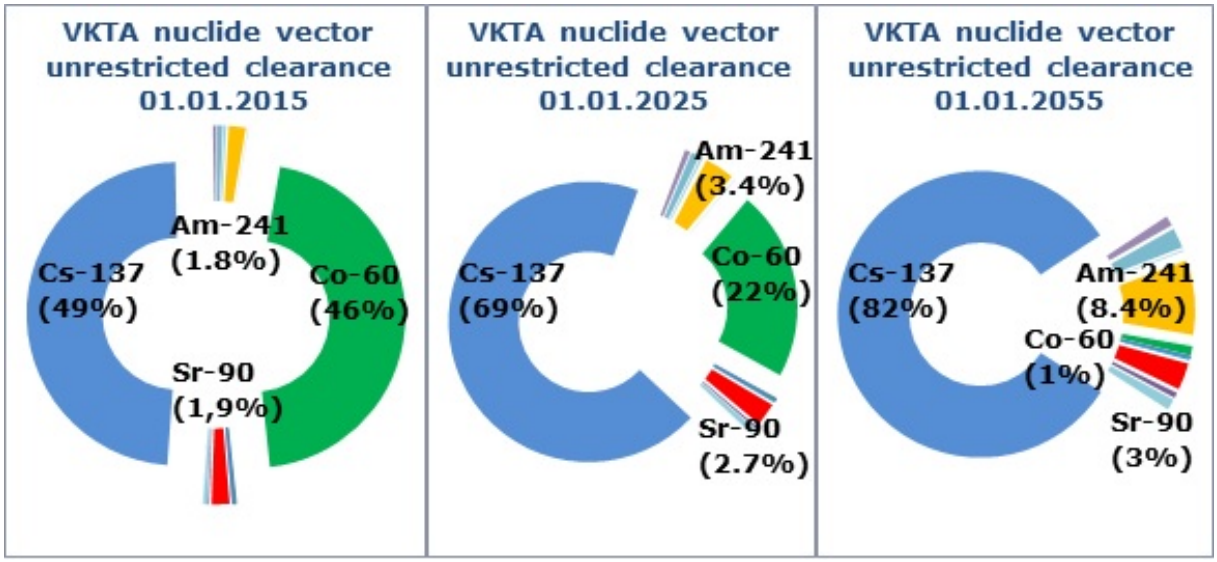

Fig. 6: Weighted and normalized VKTA nuclide vector for unrestricted clearance

We show in TABLE III and in Fig. 6 the composition of the VKTA nuclide vector, also in respect to the federal limits in Germany for unrestricted clearance (weighted NV). The calculations with HL3 however show, that even after 50 years of decay storage not a single waste bundle (Big Bag) would fulfil the federal limits for unrestricted clearance.

Thus in Fig. 7, the mass potentially releasable after decay storage is shown only for restricted clearance. For 100 Mg/a clearance and after 15 years of decay storage about 85% could be released until 2025 on basis of the sum rule, see (Eq. 1). Effectively this will be 70%, since 100 Mg/a * 15 a = 1500 Mg. Note that the shortage of disposal sites accepting waste with restricted clearance is currently one if the main obstacles for this release path in Germany.

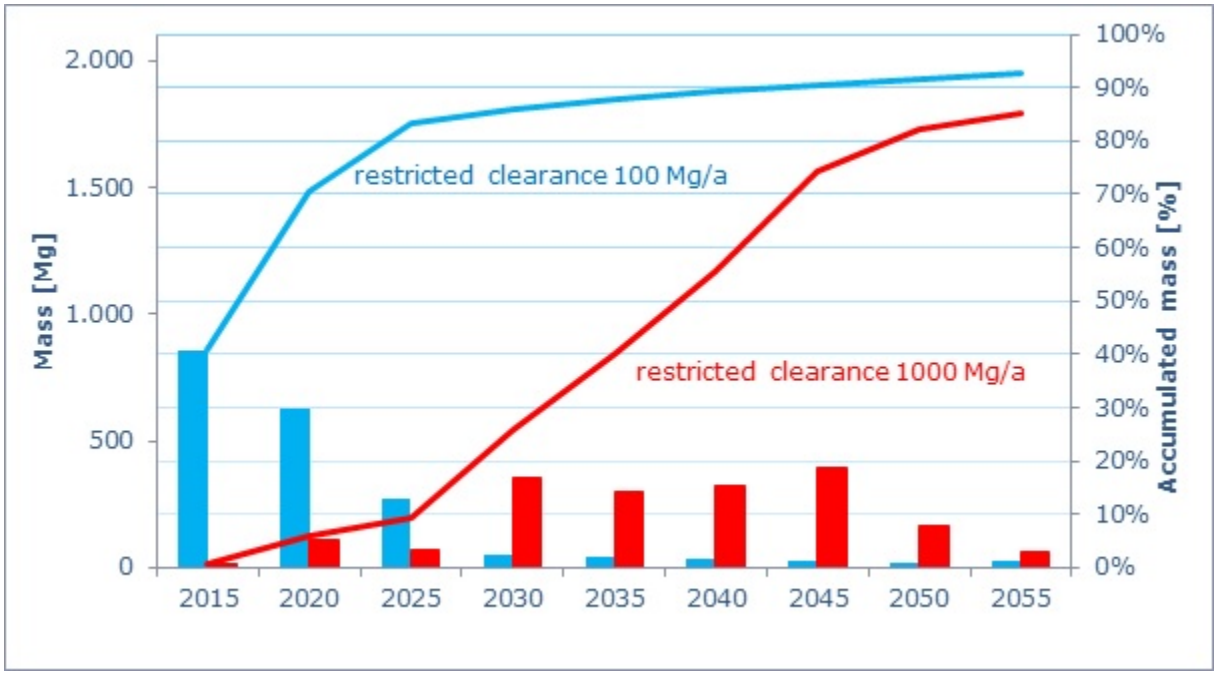

Fig. 7: Clearance after decay storage of demolition debris (VKTA nuclide vector)

Ingots from melting of metals

The dismantling process in Stade produced around 5000 Mg of metal waste, which could not be directly released, but was melted down to ingots. In this section we focus on low level metal waste melted by Siempelkamp Nukleartechnik (SNT), consisting of 877 ingots with a total mass of 888 Mg.

After the melting process, mostly Co-60 is found in the ingots while all other relevant nuclides migrate to the slag or in the gaseous phase. However, traces of Cs-137, Fe-55, Sb-125 and Nb-94 are still distributed in the ingots, but their influence on the sum value stays marginal. Thus the decay storage of the ingots are consequently mainly determined by the decline of the Co-60 inventory.

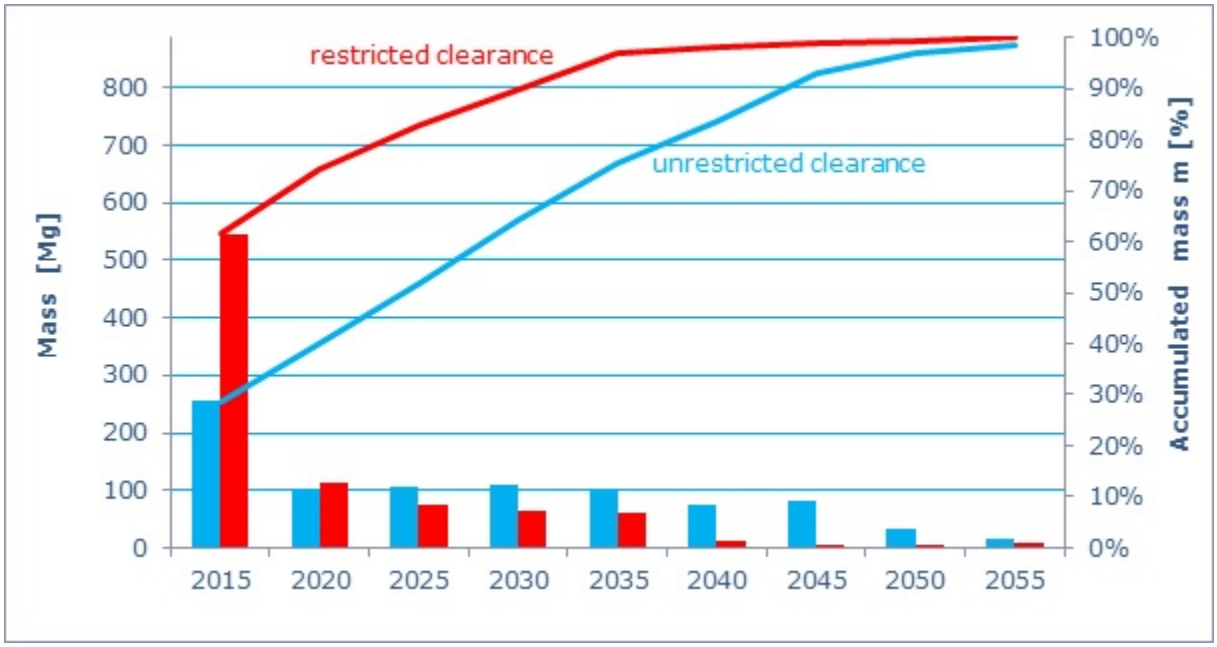

Fig. 8: Clearance potential of ingots

Fig. 8 shows that up to the year 2020 already about 40% of the here considered ingots would qualify for unrestricted clearance and 70% for restricted clearance (here metallic waste for recycling). Indeed, decay storage for ingots for a limited period of times is already implemented at the external conditioners. After melt down, the ingots are buffer stored for about 5 years. About 550 of the 877 ingots are then returned to Stade. These ingots will then indeed be released, either restricted or unrestricted. Those with higher Co-60 activity have or will be processed by the conditioner to materials used for MOSAIK containers. These are containers for intermediate level waste for final repositories.

SUMMARY AND CONCLUSIONS

We have investigated the potential of decay storage of low level radioactive waste based on the dismantling of the pressurized water reactor at the site in Stade, Germany.

For such an analysis of large amount of data we have developed the computer tool “HL3”. For given activities of the reference nuclide(s), like Co-60, and given specific nuclide vectors the tool calculates the time of decay storage for each waste entity, when the release condition is met, i.e. when the activities of the nuclides are below its corresponding federal threshold values.

In particular, our results of the evaluation of these radiologic parameters show a great potential for the decay storage of demolition rubble from concrete structures and ingots from the melting process of metals. However, due to other boundary conditions and considering a realistic implementation to the process of free release of low-level radioactive waste in Germany in Stade at the time of demolition, only a small amount of the conditioned waste in Stade could be subject to free release, on top of that amount which already has been released according to the standards and possibilities at that time.

Nonetheless, the concept of decay storage shows its theoretical potential, focusing on the radiologic parameters alone, i.e. the pure nuclide activities in relation to federal release limits. With the tool HL3 the impact of different nuclide vectors on the length of decay storage can be analyzed straight forward in a traceable and sound way. Our results thus motivate to consider in general the option of interim decay storage to reduce radioactive waste in considerable amounts in the decommissioning process of international power plants.

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