APPROACHES TO EVALUATION OF SPENT NUCLEAR FUEL REPROCESSING PRODUCTS ACTIVITY AND VOLUME EQUIVALENCE WHICH IS RETURNED TO A SUPPLIER STATE IN THE RUSSIAN FEDERATION

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Abstract

According to a series of interstate agreements of the Russian Federation, spent fuel assemblies from the Russian-origin reactors are subject to return to the Russian Federation for interim technological storage and subsequent reprocessing. Meanwhile according to the legislation of the Russian Federation, products of SNF reprocessing are subject to return to the Supplier's state. The principle of activity equivalent for the imported SNF and the reprocessing products returned to the Supplier's state is used in the Russian Federation to determine the volume of reprocessing products to be returned, taking into account the natural decay of radionuclides for the period of technological storage. However, there is no uniform approach to determine the activity equivalent criteria. The paper describes the main approaches implemented in the Russian Federation to determine this criteria and shows prospective ways to its definition.

1. INTRODUCTION

Today, there are tens of NPP power units with VVER-type reactors, such as the Akkuyu NPP (Turkey), the Belarusian NPP, the Ruppur NPP (Bangladesh) and others under construction or planned for construction overseas by ROSATOM [1]. In addition, a number of countries have been successfully operating power units with VVER-type reactors developed by Russian specialists [1]. For example, there are 4 power units with VVER-440 reactors in operation at the Paks NPP (Hungary), and two additional power units with VVER-1200 reactors are planned to be built [2]. The Kozloduy NPP (Bulgaria) also operates 2 power units with VVER-1000 reactors [3].

The nuclear fuel used at these NPPs is produced at the Russian enterprises and due to the complexity of the SNF reprocessing technology and its unavailability to foreign partners, in some cases, it is assumed that spent fuel assemblies (SFA) from reactors of the Russian-origin should be sent to the Russian Federation for its technological storage and reprocessing.

One of the main enterprises engaged commercially in SNF reprocessing in the Russian Federation is the RT-1 plant of the FSUE PO Mayak, that can reprocess SFA from VVER-440 and VVER-1000. One of the main products of SFA reprocessing is high-level radioactive waste (HLW) vitrified in the aluminophosphate matrix [4]. Table 1 presents approximate activity values of the main radionuclides included in such HLW.

TABLE 1. RADIONUCLIDE COMPOSITION OF HLW VITRIFIED IN ALUMINOPHOSPHATE MATRIX [5].

Radionuclide	Activity, Bq/l
U+Pu+Np	$1.16 \cdot 10^{10}$
Minor actinides (Am+Cm)	$9.47 \cdot 10^{10}$
$^{137}Cs+^{90}Sr$	$1.01 \cdot 10^{13*}$
Fission products (Sm, Sn, Ce etc.)	$4.89 \cdot 10^{10}$

* The activity value is given taking into account the decay products progeny (^{137m}Ba и ^{90}Y)

2. CURRENT APPROACHES TO THE RETURN OF REPROCESSING PRODUCTS TO THE SUPPLIER'S STATE

The SNF importation into the Russian Federation for the purpose of interim technological storage and/or reprocessing is carried out on the basis of the provisions of part 4 of article 48 of the Federal law of January 10, 2002 No. 7-FZ «On environmental protection» [6] and article 64 of the Federal law of November 21, 1995 No. 170 FZ «On the use of atomic energy» [7]. Under the provisions of [6, 7], the importation of SFA from foreign NPP reactors for the purpose of interim technological storage and/or reprocessing in the Russian Federation is carried out in accordance with the international agreements of the Russian Federation in the order established by the Government of the Russian Federation taking into account the basic principles of ensuring nuclear non-proliferation, environmental protection and the economic interests of the Russian Federation,, as well as the principle of priority return of radioactive waste (RW) generated after reprocessing to the state of origin of SFA.

According to requirements of the second paragraph of article 64 [7] and part 4 of article 48 [6], the Decree of the Russian Federation Government of 11.07.2003 No. 418 [8] approved the regulations on import to the Russian Federation of SFA from nuclear reactors. According to p. 17 [8], the volume of reprocessing products to be returned to the Supplier's state should be determined by the methods agreed by the parties on the basis of the activity equivalence of previously imported SFA and reprocessing products, taking into account the natural decay of radionuclides for the period of technological storage and during reprocessing. It should be noted that [8] does not contain the procedure for calculating the activity equivalent or determining the term and allows for a broad interpretation of this requirement.

Taking into account the above-mentioned variability and in accordance with the seventh paragraph of article 6 [7], the Order of Rostechnadzor of 30.12.2013 No. 655 approved the safety guide for the use of nuclear energy "Recommendations to Safety Ensuring the Return of the Irradiated Fuel Assemblies Reprocessing Products to the Supplier's State" RB-092-13 [9]. In accordance with the recommendations of this safety guidelines, reprocessing products should be sent to the Supplier's state in the form of solidified high-level radioactive waste. Illustration of the approach to manage the return of products of SNF reprocessing to the Supplier's state implemented in the Russian Federation is shown in Fig. 1.

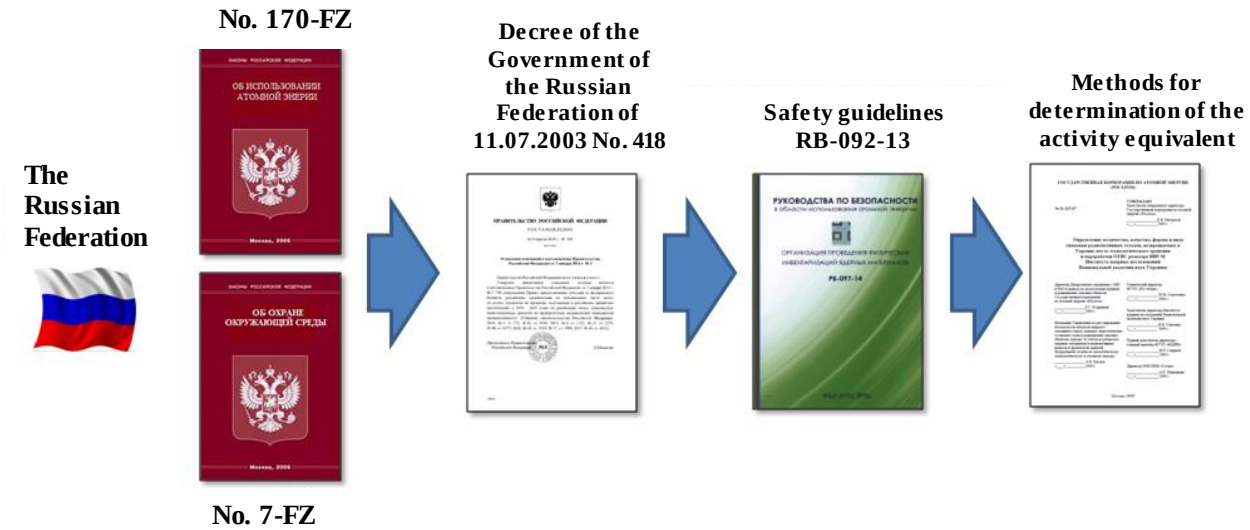

FIG. 1. Regulatory framework for the return of reprocessing products to the Supplier's state.

It is recommended to use the equality criteria of dose equivalents of SFA imported at the time of RW return and RW to be returned, taking into account the dose factors for the receipt of radionuclides with food (according to Annex2 to NRB-99/2009 [10]) for determination of the activity equivalent and the volume of RW to be returned in accordance with the provisions of RB-092-13 [9]. The dose equivalent of a single radionuclide is determined by the formula:

$$\mathrm{E}^i(t) = A^i(t) \cdot K^i \quad (1)$$

where $A^i(t)$– is the activity of the i-th radionuclide at time t;

K^i – is the dose coefficient of the i-th radionuclide.

It should be noted that during the development of RB-092-13 recommendations [9] a technological extraction of U, Pu and Np was taken into account as well as the fact that these radionuclides would remain in the Russian Federation as target products to be used in the nuclear fuel cycle. Therefore, the dose equivalents of these radionuclides are not taken into account in the calculations.

In accordance with the recommendations [9], it is necessary to use the following ratio to determine the SNF dose equivalent after a certain period from the moment of reactor core discharge:

$$\mathrm{E}^{SNF}(t) = \mathrm{E}^{FP}(t) + \mathrm{E}^{actin}(t) + \mathrm{E}^{AP}(t) \quad (2)$$

where E^{FP}, E^{actin}, E^{AP} – the dose equivalents of fission products, actinides and activation products.

The dose equivalent of vitrified HLW is defined as the sum of dose equivalents of all radionuclides in HLW:

$$E^{HLW}(t) = \sum_{i=1}^{k} E_i^{HLW}(t) \quad (3)$$

In most cases, vitrified HLW is understood to be HLW in the aluminophosphate matrix, the approximate radionuclide composition of which is shown in Table 1. However, the radionuclide composition of HLW may change along with the development of vitrification technologies.

The dose equivalent of HLW to be returned should be equal to the dose equivalent of SNF imported at the time of return:

$$E^{HLW}(t_{return}) = E^{SNF}(t_{return}) \quad (4)$$

This approach to determination of the activity equivalent of SFA and RW correlates with the approaches used in international practice. Thus, the Sellafield Ltd (UK) uses «toxic potential» as a criteria of the activity equivalent of radioactive substances with different radionuclide composition [11]. According to the definition given in [11], the toxic potential of radioactive material means the volume of water in which this radioactive material must be completely dissolved to such an extent that the expected dose from the consumption of water by population from this source during the year does not exceed 1 mSv/year. This approach to determining the activity equivalent of various radioactive materials aimed at comparison of the hypothetical dose effects on the population of all radionuclides contained in them when they enter the body and correlates with the approach implemented in RB-092-13 [9].

Today, there is a wide practice of using RB-092-13 [9] in determining the amount of RW returned to the Supplier's state, including the imports of SNF from research reactors of Russian-origin.

3. NEW POSSIBLE APPROACHES TO THE RETURN OF REPROCESSING PRODUCTS TO THE SUPPLIER'S STATE

The provisions of RB-092-13 [9] should be regarded as recommendations and, according to paragraph 5 [9], the mandatory requirements of regulatory documents can be met with the use of other approaches with the validity of such approaches to ensure safety. RB-092-13 [9] recommendations were developed on the basis of an ideology that did not provide for the return to the Supplier's state of such products as nuclear fissile materials (NFM) – regenerated uranium (RepU), plutonium (Pu), neptunium (Np). Therefore, these products are not taken into account when determining the dose equivalent of RW to be returned. It is assumed that NFM are valuable products and will be used in the Russian Federation in the form of regenerated nuclear fuel (e.g. MOX fuel).

Nevertheless, an approach is currently being worked out in the Russian Federation providing the return of products of reprocessing to the Supplier's state in the form of fresh nuclear fuel containing regenerated NFM. In this case, there can be a significant reduction in the amount of HLW returned to the Supplier's state as a result of taking

into account the dose equivalent of regenerated NFM, included into the fresh fuel assemblies. This fact can deliver the Supplier's state from the necessity of the construction of large-scale storage and/or HLW repositories in deep geological formations on its territory which is prescribed by paragraph 1.17 of General safety requirements IAEA No. GSR Part 5 [10].

It should be noted that the export of regenerated nuclear materials does not contradict the international obligations of the Russian Federation on non-proliferation of nuclear weapons [12], as these materials are supplied in the form of fresh fuel assemblies. In addition, such option for the return of reprocessing products is implemented, for example, at the Borssele NPP (the Netherlands), which includes a single PWR-type reactor [13]. SNF generated during the operation of this reactor is sent for processing to the La Hague plant (France), and the equivalent of reprocessing products is returned to the Netherlands, including in the form of MOX fuel. At the same time, the power unit of this NPP during the first forty years of its operation was loaded with uranium oxide nuclear fuel manufactured according to classical technologies [13].

In practical terms for the Customers of the SNF reprocessing services, the above approach will lead to:

— a significant reduction of the volume of HLW to be returned, in case the Suppliers' states use regenerated NFM in their NFC;
— full compensation of the HLW activity and its staying in the Russian Federation, in case the Suppliers' states use regenerated NFM in their NFC in quantities larger than imported SNF can contain.

Today, the Russian Federation has accumulated experience in the fabrication of nuclear fuel containing regenerated nuclear materials. For example, fuel rods with REMIX fuel being operated in the VVER-1000 reactor of unit 3 of the Balakovo NPP [14]. The basis for REMIX fuel is the undivided mixture of uranium dioxide and plutonium, formed during the reprocessing of SNF from VVER reactors after its purification from other actinides and fission products. Enriched uranium is added to this mixture to create REMIX fuel. In addition, the experimental-industrial operation of the MOX-fuel production plant is being carried out at the FSUE MCC [15].

The results of a preliminary analysis of products of reprocessing to be returned to the Supplier's state considering of the return of regenerated nuclear materials in the form of fresh nuclear fuel are described below., In addition, a comparison of this option with the current approach was carried out. The results of calculation of the nuclide activity of VVER-1200 SNF with initial enrichment of 4.95 % 235U and a burn-up of 60 GWd/tHM were used as initial data for analysis. The calculations were performed with the use of generally recognized code SCALE [16]. The composition of vitrified HLW (see Table 1), as well as fresh REMIX and MOX fuel [17, 18], were also used as a initial data for the analysis considering the dose equivalents of the corresponding radionuclides.

It was assumed that prior to delivery to the Russian Federation, the spent fuel is stored for 10 years at foreign NPP, and the return of reprocessing products is carried out through 10 years after delivery of SFA to the Russian Federation (20 years after the reactor core discharge). The change in SNF activity during this period of time caused by radioactive decay of various nuclides (Fig. 2) is taken into account.

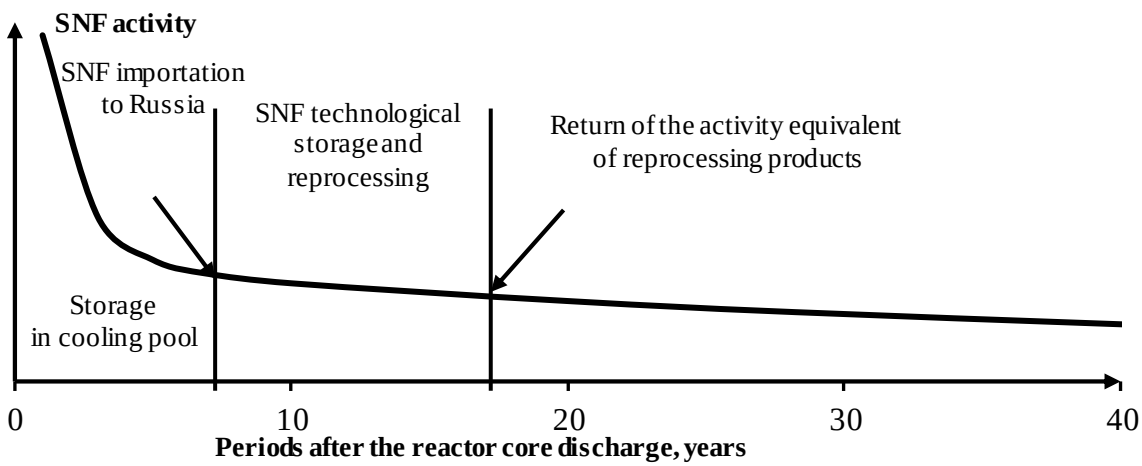

FIG. 2. Typical SNF activity dependence on the time and the main stages of SNF management.

The following ratio was used to determine the SNF dose equivalent after a certain period from the moment of reactor core discharge:

$$E^{SNF}(t) = E^{FP}(t) + E^{actin}(t) + E^{AP}(t) + E^{TP}(t) \quad (5)$$

where E^{FP}, E^{actin}, E^{AP}, E^{TP} – The dose equivalents of fission products, actinides, activation products and target products (regenerated nuclear materials).

The dose equivalent of fresh fuel containing regenerated nuclear materials (REMIX- and MOX-fuel) is determined by the formula:

$$E^{FA}(t) = E^{238}(t) + E^{239}(t) + E^{240}(t) + E^{241}(t) + E^{242}(t) + E^{*}(t) \quad (6)$$

where E^{238}, E^{239}, E^{240}, E^{241} and E^{242} – the dose equivalents of isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu;

E^{*} – the dose equivalents of isotopes ^{232}U и ^{236}U (for REMIX fuel) or the dose equivalent of isotope ^{241}Am (for MOX fuel).

The dose equivalent of HLW is determined by the formula (3).

In case of implementing the activity equivalent principle for one imported SFA and fuel assembly containing reprocessed nuclear material (REMIX or MOX fuel) and vitrified HLW, the HLW mass for a single SFA (MHLW) is determined by the formula:

$$M^{HLW} = \frac{E^{SFA} - E^{reg}}{E^{HLW}} \quad (7)$$

where E^{SFA}, E^{HLW} и E^{reg}г – the dose equivalents of one SFA, vitrified HLW and fresh fuel assemblies containing regenerated nuclear materials.

Table 2 shows the results of the calculations of the HLW mass, for the technology of their inclusion into the aluminophosphate matrix, and to be returned under different scenarios.

The data presented in Table 2 show that the return of the dose equivalent of imported SNF in the form of fresh fuel containing regenerated nuclear materials leads to a significant (up to 8 times) reduction in the amount of returned RW.

TABLE 2. WEIGHT OF VITRIFIED HLW TO BE RETURNED (WHEN IMPORTING ONE SFA CONTAINING 450 KG OF NUCLEAR FUEL).

Scenario of return	HLW mass to be returned		
	Return of HLW only (with the dose equivalent of NFM in SNF)	Return of HLW and one fuel assembly with REMIX fuel	Return of HLW and one fuel assembly with MOX fuel
1	2	3	4
HLW mass, kg	2128	1848	264

The possibility of HLW fractionation with the separation of the fraction, the activity of which is formed by the isotopes of caesium and strontium (making the main contribution to the activity of HLW) is regarded as an additional direction of the development of SNF reprocessing technology and HLW management in the Russian Federation. Along with the introduction of the technology of HLW vitrification into the borosilicate matrix, due to the concentration of greater activity (up to 1000 Ki/kg [19]) in a small volume, such an option will allow:

— to form a more compact HLW matrix;
— to abandon the construction of repositories for final disposal in deep geological formations.

The generation of vitrified HLW containing only caesium and strontium has not been carried out commercially in the Russian Federation yet. Due to that fact, the exact radionuclide composition of the HLW was not determined. However, a preliminary assessment of the nuclide composition of vitrified HLW can be obtained on the assumption that all activity of the RW vitrified into the borosilicate matrix is caused only by isotopes 137Cs and 90Sr, as well as by their decay products. Other radionuclides are absent in the composition of the matrix. A preliminary assessment of the HLW activity is presented in Table 3.

TABLE 3. RADIONUCLIDE COMPOSITION OF THE HLW VITRIFIED INTO BOROSILICATE MATRIX.

Radionuclide	Activity, Bq/l
^{137}Cs	$4{,}32\ 10^{13*}$
^{90}Sr	$5{,}76 \cdot 10^{13*}$

* The activity value is given taking into account the decay products progeny (^{137m}Ba and ^{90}Y)

As part of this analysis, a preliminary calculation of the volume of reprocessing products to be returned to the Supplier's state was made, taking into account the return of the SNF activity equivalent in the form of HLW with isotopic composition specified in Table 3. Similar to the above calculations, it was assumed that the initial enrichment of SNF by 235U is 4.95 %, and the burnout depth is 60 GWd/tHM. Table 4 shows examples of calculations of the masses of fractionated HLW vitrified into borosilicate matrices to be returned to the Supplier's state, including scenarios for the return of fresh nuclear fuel containing regenerated nuclear materials.

TABLE 4. MASS OF FRACTIONATED HLW VITRIFIED INTO BOROSILICATE MATRICES TO BE RETURNED (WHEN IMPORTING ONE SFA CONTAINING 450 KG OF NUCLEAR FUEL) [19].

Scenario of return	HLW mass to be returned		
	Return of HLW only (with the dose equivalent of NFM in SNF)	Return of HLW and one fuel assembly with REMIX fuel	Return of HLW and one fuel assembly with MOX fuel
1	2	3	4
HLW mass, kg	667	580	87

The above results show that in case of the return of HLW only, its volume significantly reduced due to the implementation of the fractionation compared with unfractionated HLW (see Table 2). At the same time, when returning the activity equivalent in the form of fresh MOX-fuel together with fractionated HLW, their amount may be less than 100 kg per one SFA imported to the Russian Federation for technological storage and subsequent reprocessing.

4. CONCLUSION

The approaches implemented in the Russian Federation to the return of reprocessing products to the Supplier's state are based on the analysis of dose coefficients of various nuclides and correspond to international approaches. Nevertheless, the Russian approach can be further improved through the involvement of the regenerated nuclear materials.

Despite the fact that the precise volumes of reprocessing products to be returned can be determined only on the basis of the SNF specific characteristics (burn-up, initial enrichment, the mass of nuclear fuel, exposure time, etc.), the results of preliminary analysis show that the volume of RW to be returned to the Supplier's state is significantly reduced in case of the return of the SNF activity equivalent in the form of fresh uranium-plutonium fuel (REMIX - or MOX-fuel). In addition, the volume of returned radioactive waste is significantly reduced when it is vitrified into the borosilicate matrix (only caesium and strontium are included).

REFERENCES

[1] Official website of the state atomic energy Corporation "Rosatom" [Electronic resource]. URL: http://www.rosatom.ru/.

[2] Paks NPP Official Site [electronic resource]. URL: http://www.atomeromu.hu/en/.

[3] Kozloduy NPP Official Site [electronic resource]. URL: https://www.kznpp.org/.RESEARCHER, A., Organization, unpublished data.

[4] "Plans of SNF reprocessing at FSUE PO Mayak 2030", the CEG Workshop, IAEA "Economics of using spent fuel: reprocessing and direct isolation", Stockholm, (2011).

[5] REMIZOV, M. B., "The development of methods for the characterization of vitrified HLW FSUE, PO Mayak for the transfer of FSUE NO RWM for disposal in geological formations", Scientific-technical seminar "Problems of processing and conditioning of RW when rendering to the eligibility criteria", Saint-Petersburg (2018), 4 – 9 June

[6] Federal law No. 7-FZ of 10.01.2002 (as amended on 31.12.2017) "On environmental protection".

[7] Federal law of 21.11.1995 no. 170-FZ (ed. from 03.07.2016) "On the use of atomic energy".

[8] Resolution of the Government of the Russian Federation from 11.07.2003 no 418 (edited on 04.09.2012) on approval of the regulation «On the procedure of import into the Russian Federation of irradiated fuel assemblies of nuclear reactors».

[9] Guidelines for safety in the use of nuclear energy «Safety provision recommendations at returning the processed products of irradiated heat-generating assemblies into the country of their supplier» RB-092-13, approved by the Order of the Federal service for environmental, technological and nuclear supervision of 30.12.2013 no 655.

[10] Sanitary rules and standards SanPiN 2.6.1.2523-09 "Radiation safety Standards" NRB-99/2009, approved by the decree of the Chief state sanitary doctor of the Russian Federation from 07.07.2009 no 47.

[11] HAGGER, R., GARNER, D. S. J., BEAUMONT, D. M., HESKETH, K.., Uranium oxide recycling to give more sustainable power generation, IAEA-SM-357/55.

[12] Treaty on the non-proliferation of nuclear weapons, Approved by United Nations General Assembly resolution 2373 (XXII) of 12 June (1968).

[13] National Report of the Kingdom of the Netherlands for the Sixth Review Meeting, Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management (2017).

[14] BALAKOVO NPP: innovative REMIX fuel [Electronic resource] is loaded into the reactor of unit 3 // Russian nuclear community. URL: http://www.atomic-energy.ru/news/2016/07/04/67233.

[15] Production of MOX fuel [Electronic resource] / / official website of FSUE MCC. URL: https://www.sibghk.ru/.

[16] SCALE Code System-Oak Ridge National Laboratory. Reactor and Nuclear Systems Division – ORNL/TM-2005/39, Version 6.2, April (2016).

[17] MAKOVSKY, S.V., KURYNDIN, A.V., KIRKIN, A. M., SINEGRIBOV, S.V., Analysis of the radiation and thermal characteristics of the spent REMIX fuel. Nuclear Science and Technology. Series: Nuclear reactor constants **4** (2017), 134-143.

[18] YOUINOU. G., VASILE, A., Plutonium Multirecycling in Standard PWRs Loaded with Evolutionary Fuels, Nuclear Science and Engineering **151** (2005) 25-45.

[19] Specification by AREVA NC on intermediate level vitrified waste (CSD-B) from the reprocessing of German LWR fuel elements, Statement of the Nuclear Waste Management Commission (ESK) (2010).

[20] INTERNATIONAL ATOMIC ENERGY AGENCY, Radioactive waste management before disposal, General safety requirements no. GSR Part 5, IAEA, Vienna (2010).