TRANSPORTATION OF ENRICHED REPROCESSED URANIUM

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ABSTRACT

In the last few years the volume of enriched reprocessed Uranium (RepU) transportation increased considerably and became an important issue. While the transport of enriched commercial grade Uranium is standard practice since decades the transport of RepU presents new challenges.

Reprocessed Uranium is specified in ASTM C 996 [1]. The limits given there for fission products and actinides are sufficiently low, however the limit specified for ^{232}U of 0.05 µg/gU leads to considerable consequences.

The A_2-values of ^{232}U and its decay product ^{228}Th are rather small and a concentration of roughly 1 % of the limit specified in ASTM C 996 will exceed the radioactivity allowed in type A packages which are used for the transport of commercial grade Uranium. A possible solution to this problem could be the declaration of RepU as LSA-II and the use of IP-2 packages containing fissile material. However, in some countries it is not allowed to approve IP-2 packages for fissile material. Hence, the consequence is to use a type B(U)F package.

A further problem is the decay of the aforementioned ^{232}U. In the decay chain there are nuclides with a high gamma yield at high energies, especially the nuclide ^{208}Tl. The gamma source intensity of fresh RepU increases in one year by a factor of roughly 200, in 2 years by a factor of 350, and in 10 years by a factor of 600!

This phenomenon is treated quite differently by different competent authorities. One certificate of package approval contains a rather restrictive limit for ^{232}U which ensures that the limits for the dose rates are kept at all times. Other certificates specify no limit for ^{232}U or restrict the time between reprocessing/ enrichment and transport. In this case, even if the limit given in ASTM C 996 is not exceeded, the dose rates might exceed the limits specified in the Regulations by a factor of up to 3.

This paper will illustrate the challenges mentioned and discuss the consequences for transportation of RepU.

Key words: reprocessed Uranium, transport of UF_6, shielding analysis

INTRODUCTION

Enriched commercial grade uranium hexafluoride (UF_6) is transported since decades in type A packages for fissile material which consist of 30B cylinders as primary containment and protective structural packagings (PSPs) providing mechanical and thermal protection. The 30B cylinders are specified in the international standard ISO 7195 [2], or the US standard ANSI N14.1[3]. For international shipments three designs of PSPs are currently in use.

The most important issues concerning the transport of enriched commercial grade UF_6 are criticality safety and its significant chemical hazard. To address these issues the 30B cylinder must fulfill certain leak tightness criteria. But neither the radioactivity of the content nor the dose rates at the package are important issues. The A_2 value of enriched commercial grade uranium (max. 5 wt.%

enrichment) is unlimited and the dose rates are well below the limits specified in the Regulations. These conditions change slightly after multiple refilling of cylinders containing heels. Although the A_2 value of the decay products which are concentrated in the heels is not unlimited, the total radioactivity in the package remains well below 1 A_2. The dose rate at a package loaded with a cylinder containing heels quantities of UF_6 is expected to be considerably higher than the dose rate to be expected on a filled cylinder, but is still well below the limits specified in the Regulations.
In the last few years the transport of enriched reprocessed uranium (RepU) became more and more important. For this material physically the same packagings are used as for enriched commercial grade uranium. However, these packagings loaded with RepU cannot be transported as type A packages but require type IF or type B(U)F approvals:

- The A_2 value of RepU is not unlimited but must be determined by using the mixture formula as defined in para. 405 of the Regulations
- The radioactivity of RepU in a 30B cylinder will in general exceed 1 A_2
- Dose rates at the package are much higher than for enriched commercial grade UF_6 and might exceed the limits given in the Regulations
- Heels require special attention due to the concentration of the decay products in the small quantity of UF_6 remaining in the cylinder after emptying.

The following presentation will address these issues in more detail. First, the main differences in the nuclide composition of enriched commercial grade and reprocessed uranium are shown. Then, the influence of the most important nuclide ^{232}U and its decay products on radioactivity and dose rates is discussed. An evaluation of currently and future package designs with respect to the challenges of the transport of RepU concludes the presentation.

NUCLIDE COMPOSITION OF ENRICHED REPROCESSED URANIUM [1]

The nuclide compositions of enriched reprocessed and commercial grade (natural) uranium are listed in Table 1. The comparison of the concentration of the uranium nuclides ^{232}U, ^{234}U and ^{236}U and ^{99}Tc is shown in Figure 1. The table and figure show that the concentration of ^{232}U in enriched reprocessed uranium might be 500 times higher than in commercial grade uranium.

Table 1. Nuclide composition of enriched reprocessed and commercial grade uranium

Nuclides	Unit	Enriched reprocessed uranium	Commercial grade uranium
U-232	wt. % U	5.0×10^{-6}	1.0×10^{-8}
U-234	wt. % U	2.0×10^{-1}	5.5×10^{-2}
U-236	wt. % U	3.0×10^{0}	2.5×10^{-2}
Fission products	MeVBq/kgU	4.4×10^{5}	-
Tc-99	wt. % U	5.0×10^{-4}	1.0×10^{-6}
Neptunium and plutonium	Bq/kgU	3.3×10^{3}	-

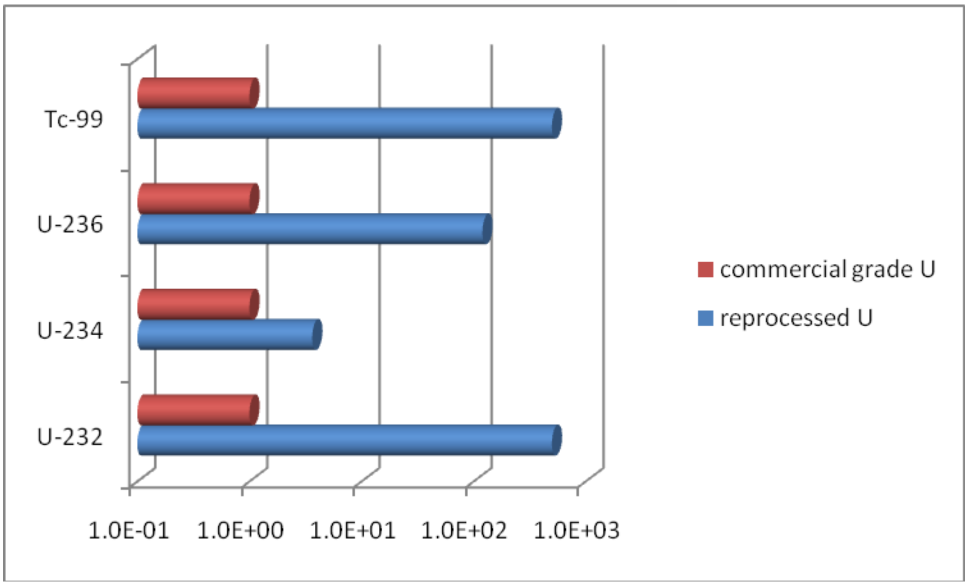

Figure 1. Comparison of the nuclide composition of enriched reprocessed with commercial grade uranium (logarithmic scale for ratio RepU/commercial grade)

DECAY CHAIN OF ^{232}U AND RADIOACTIVITY OF REPU

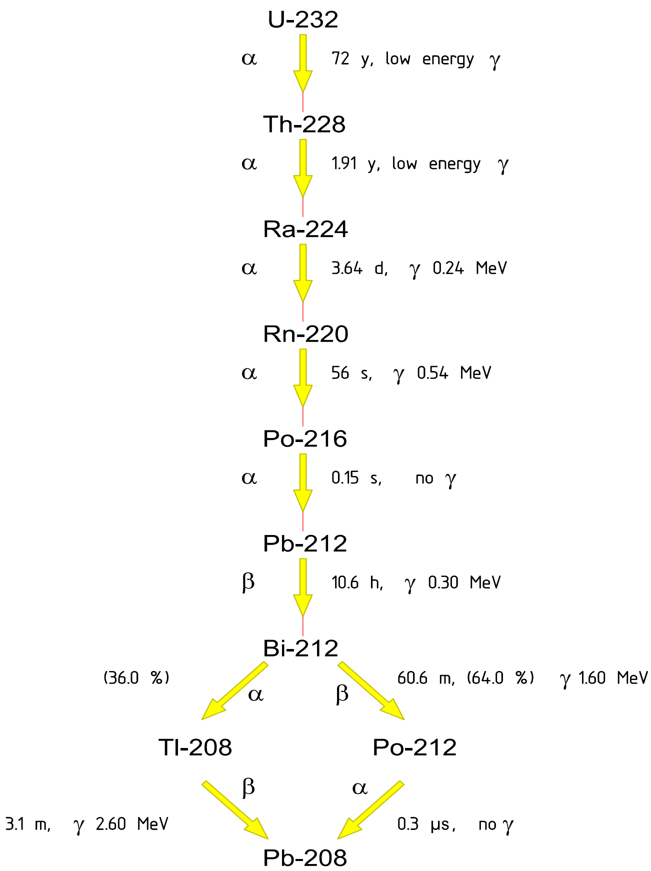

Figure 2. ^{232}U decay chain

Decay chain of ^{232}U

The decay chain of ^{232}U is part of the Thorium decay chain and is shown in Figure 2.

Radioactivity of filled cylinders

The main nuclides relevant for the determination of the radioactivity in A_2 of enriched reprocessed uranium are ^{232}U with the decay product ^{228}Th and ^{234}U. The A_2 value of ^{232}U does not contain the contribution of ^{228}Th, but the A_2 value of ^{228}Th contains the contributions of all decay products in the decay chain shown in Figure 2.

Figure 3 shows the development of the radioactivity over a time period of 10 years. The contribution of ^{234}U is constant over this period with in total about 7.9 A_2. ^{232}U decays slightly (half-life 72 years) from an initial value of 6.3 A_2, and the contribution of the decay product ^{228}Th increases and reaches after 10 years about 57 A_2. The total radioactivity in a 30B cylinder containing enriched reprocessed uranium is maximal approx. 70 A_2. Assuming a homogeneous distribution of this activity in a quantity of 2277 kg UF_6 leads to a specific activity of 3×10^{-5} A_2/g which is well below the limit for LSA-II. Thus, the package could be transported either as type IF or as type B(U)F package.

Radioactivity of cylinders containing heels

During emptying of a cylinder at the destination an unknown amount of the impurities and decay products of uranium remain in the cylinder. Assuming conservatively that most of the decay products remain in the heels the condition for the specific activity of LSA-II is not met anymore and a type B(U) package is required.

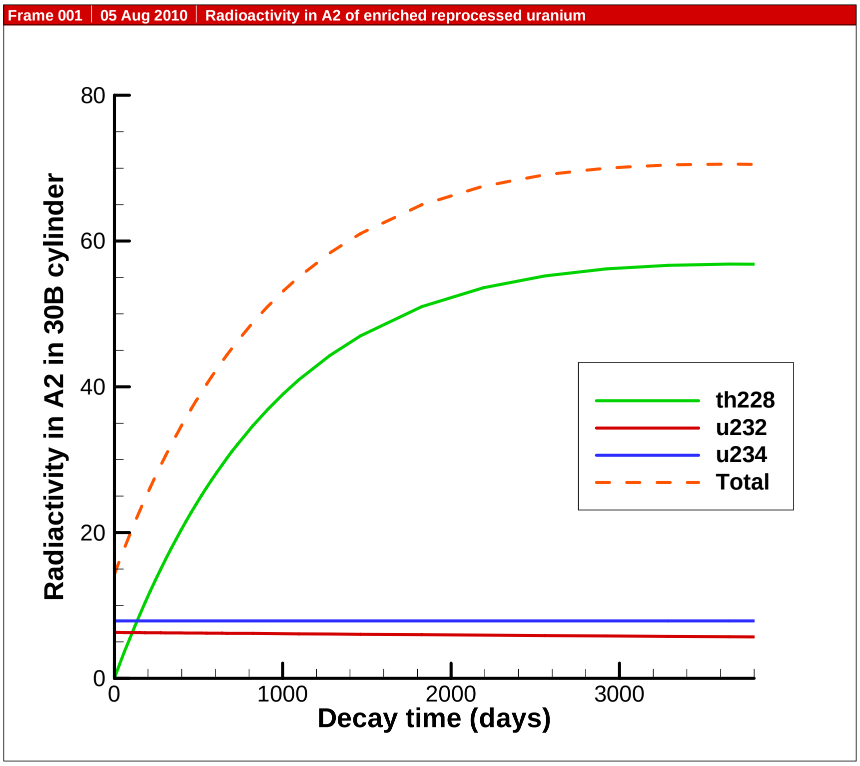

Figure 3. Total radioactivity in a 30B cylinder filled with enriched reprocessed uranium

GAMMA RADIATION SOURCE TERM AND DOSE RATES

Gamma radiation source term

At the end of the decay chain of ^{232}U shown in Figure 2 ^{208}Tl decays to the stable nuclide ^{208}Pb, emitting hard gamma radiation with intensity of about 100% and energy of 2.6 MeV. The concentration of ^{228}Th and its decay products increases in equilibrium for about 10 years and decreases then with the parent ^{232}U.

Figure 4 shows the development of the gamma source intensity over a time period of 10 years. The contribution of ^{232}U increases considerably with time and is after 10 years more than a factor of 10 higher than the contribution of all other nuclides. The contributions of ^{234}U and ^{236}U are constant over this period. The contribution of ^{238}U increases for about 3 months because of its decay products and remains then constant. The contribution of the fission products with ^{137}Cs selected here as representative is only relevant for recently processed material and is of minor importance after a relatively short storage and/or transport time.

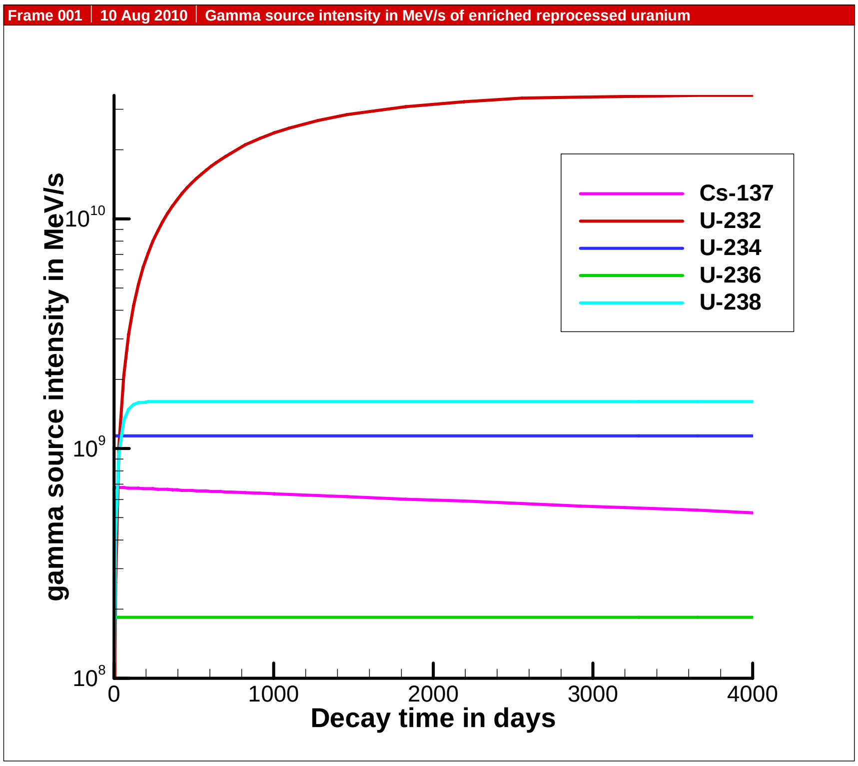

Figure 4. Gamma source intensity in a 30B cylinder filled with enriched reprocessed uranium

<u>Dose rates at a typical PSP loaded with a filled cylinder</u>

The gamma dose rates at a typical PSP increase considerably with time. Figure 5 shows that dose rates for RepU are sufficiently small for short times between processing and transport (separation of ^{228}Th). With increasing time after processing the dose rates increase by factors.

The dose rate at the surface of a typical PSP remains well below the limits of the Regulations. However, the dose rates in 2 m distance from the vehicle might exceed the limits of the Regulations after the accumulation of storage period before transport and transport time reaches a certain limit.

The most important values are the dose rates in 1 m distance from the surface of the package. After about 1.5 years after reprocessing this value exceeds 100 µSv/h which means that the transport index exceeds TI=10. Therefore it might happen that a transport leaves the consignor under not exclusive use conditions but it should arrive at the destination under exclusive use conditions.

<u>Dose rates at a typical PSP loaded with a cylinder containing heels</u>

As discussed above the decay products of uranium are concentrated in the heels after emptying. Depending on the assumed scenario – concentrated bottom pool or contamination of the inner surface of the cylinder – higher dose rates at the surface or in 2 m distance from the vehicle must be expected than for the filled cylinder. It might be required to store the cylinder for some time to allow decay of the nuclide ^{228}Th and decrease of dose rates below the limits specified in the Regulations. In any case transport under exclusive use might be necessary.

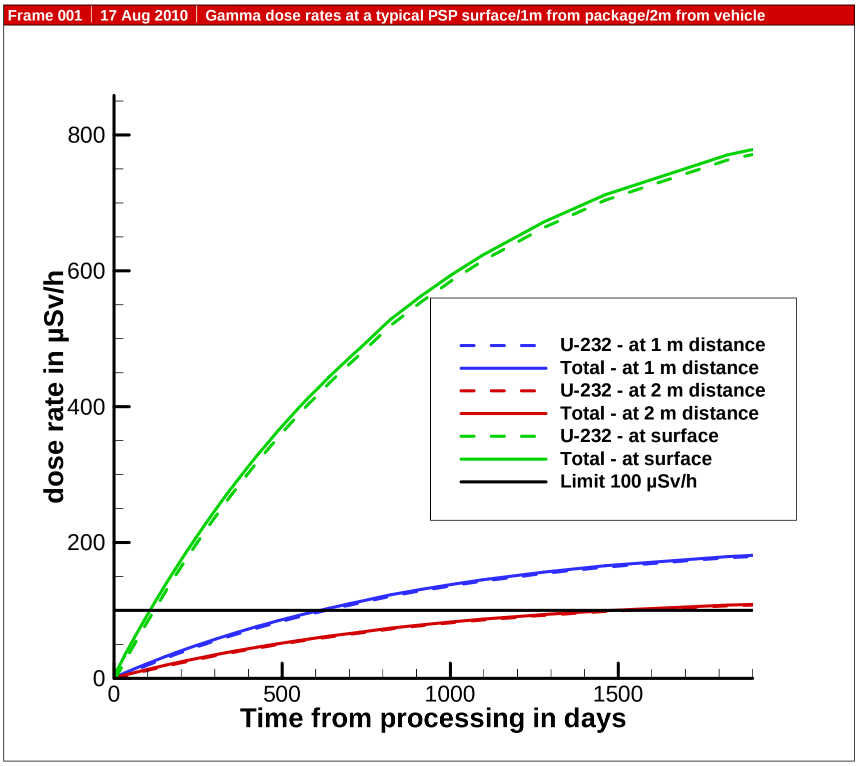

Figure 5. Dose rates at a typical PSP

CURRENT DESIGNS AND THEIR LICENSING CONDITIONS

Currently, there are three package designs in use for international transports. A new package design is under development and presented in another paper given during this PATRAM [4]. The following evaluation will concentrate on the technical and administrative aspects relevant for transportation of RepU but will not point out any other details of the different package designs.

<u>Design A (Type IF)</u>

The specification of RepU mentioned in the certificate of package approval of design A is in principle based on the ASTM C 996[1]. However, for some nuclides lower values are specified than given in [1], e. g. for the nuclide ^{232}U only 20% of the concentration given in [1] is permitted. Additionally, the permissible activity of the decay products of uranium 2 years after filling of the cylinder is specified. There are no specific requirements for the transport of heels.

<u>Design B (Type IF, B(U)F)</u>

The specification of RepU mentioned in the certificate of package approval of design B is based on the requirements for LSA-II given in the Regulations. There are no restrictions for the concentration of the nuclide ^{232}U or the activity of the decay products of uranium as function of time. However, the amount of the fissile nuclide ^{233}U is restricted to a low value.

<u>Design C (Type B(U)F)</u>

The specification of RepU mentioned in the certificate of package approval of design C is partially based on ASTM C 996 [1]. The limits for fission products and alpha activity of neptunium and plutonium are explicitly specified exactly like in [1]. No limits are given for ^{232}U, ^{234}U and the decay products of uranium. The total activity is limited to $10^5 A_2$. There are no specific requirements for the transport of packages loaded with cylinders containing heels.

New Design presented in [4] (Type IF, B(U)F)

The specification of RepU mentioned in the application for the new design presented in [4] is fully based on ASTM C 996 [1]. In addition to that also a specification of RepU exceeding [1] is permitted. For type IF packages a total permissible radioactivity of 227 A_2 is specified which complies with the permissible specific activity for LSA-II multiplied by the maximal permissible mass of UF_6 in a 30B cylinder. This activity limit is kept for type B(U)F packages as well. Special care has been taken for the specification of the permissible concentration of ^{232}U as function of the time between filling of the cylinder and transport to avoid that the dose rates at the package and vehicle exceed the relevant limits at the end of the transport, taking into account an envelope transport time of 1 year. For the transport of heels special requirements are specified to take into account higher concentration of the decay products and the expected higher dose rates.

CONCLUSIONS

The transport of enriched reprocessed uranium in the form of UF_6 seems to be a real challenge for the designers of packages as well as for the Competent Authorities.
The challenge for the designers are the properties of ^{232}U. Against the usual expectations there is no decrease of radioactivity and dose rates with time. Instead, an increase of radioactivity and dose rates by factors could occur depending on the combination of filling date, start of transport date and arrival date. Therefore, a thorough analysis of acceptable concentrations of ^{232}U depending on the time period between filling of the cylinder and arrival at the destination is necessary to ensure compliance with the Regulations from start to end of transport. Another challenge for the designers of packages are empty packages containing heels. Against expectations again, the dose rates at a package containing heels quantities of UF_6 could be much higher than the dose rates at a package containing a filled cylinder. Depending on the initial concentration of ^{232}U and the time period between filling and emptying intermediate storage of the empty cylinders might be required before their transport in a PSP in compliance with the Regulations might be possible.
The challenge for the Competent Authorities is certainly to find a common approach to the specification of RepU in certificates of package approval. The content specifications in the three existing approvals differ significantly. In two of the approvals there is no limit on the ^{232}U concentration. The permissible radioactivity of $10^5 A_2$ (equivalent to several 100 g of plutonium) specified in one of the certificates appears to be quite high for such a kind of package design. In two of the approvals the transport of packages containing heels is allowed but not regulated separately and in the third approval transport of packages containing heels is not permitted. The specification of a maximum concentration of ^{233}U is unique in one approval.
The validation of the certificates of package approval in other countries than the country of origin complicates the current situation by far. Type B(U)F approvals are validated in some countries only as type AF or as special arrangement. In some validations new limits for the minimum permissible mass of UF_6 are introduced and/or cylinders containing heels are excluded. Some validations or special arrangement have other validity periods than the original certificate of package approval.
With the application for the new design presented in [4] a content description was submitted to the Competent Authority which envelopes the different approaches currently on the market and

addresses the increase of activity and dose rate with time in a safe but also economic way. Furthermore, the content description contains conditions for the transport of heels which allow its transport meeting the Regulations in full.

REFERENCES

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