

**EXTERNAL DOSE RATE ANALYSIS OF THE NEW DN30 PACKAGE FOR THE
TRANSPORT OF UF₆**

Maik Hennebach, Franz Hilbert

DAHER NUCLEAR TECHNOLOGIES GmbH, Hanau, Germany

ABSTRACT

The DN30 package was developed by DAHER NUCLEAR TECHNOLOGIES GmbH (DAHER NT) for the transport of enriched commercial grade and reprocessed UF₆ up to an enrichment of 5 %. It consists of a standard 30B cylinder and the DN30 Protective Structural Packaging (PSP) and is licensed as a type AF, IF and B(U)F package. The design and several aspects of the license process were already presented at PATRAM 2013 and 2016.

The present paper is focused on UF₆ contents from reprocessed uranium, where the external dose rates are dominated by the source term contribution from daughter nuclides of ²³²U, especially ²⁰⁸Tl as a hard gamma emitter. Our covering assessment of the source term takes into account the initial ²³²U content, the build-up and decay of uranium daughters as well as the contributions from impurities like higher actinides and fission products.

For 30B cylinders containing heels, further complexity is added to the analysis: the amount of radionuclides remaining in the heels after emptying needs to be assessed as well as the impact of the position and distribution of the heels on the external dose rates. In general, dose rates to be expected at the surface of the package and in 2 m distance from the vehicle are higher for cylinders containing only heels than for full cylinders and might exceed the dose rate limits of the Regulations for some time after emptying.

The paper will present an overview of our analysis methods and results for the dose rate assessment of full cylinders and cylinders containing heels. Furthermore, a short outlook on potential solutions for heels cylinders with excessive dose rates is presented.

INTRODUCTION

The DN30 is licensed as a type AF, IF and B(U)F package for the transport of enriched commercial grade and reprocessed UF₆ with an enrichment of up to 5 wt.%. The dose rate analysis needs to take the source term contribution from uranium decay products into account, as well as contributions from impurities in the UF₆ content, i.e. higher actinides and fission products.

OVERVIEW OF THE DN30 PACKAGE

The DN30 package consists of the 30B cylinder, as specified in the standards ISO 7195 [1] and ANSI N14.1 [2], and the DN30 PSP. The DN30 PSP accommodates the 30B cylinder and provides the mechanical and thermal protection for the 30B cylinder during RCT, NCT and ACT. The DN30 PSP consists of a top and bottom half which are connected by a closure system consisting of 6 individual closure devices, 3 on each side of the PSP. A gasket is fitted on the step-joint part between both halves to prevent water inleakage.

The halves consist of an inner and outer stainless steel shell as well as energy-absorbing and insulating closed-cell PIR foam (fire retardant PU foam) of different densities filling the space between the shells.

An overview of the major components of the DN30 package is shown in Figure 1.

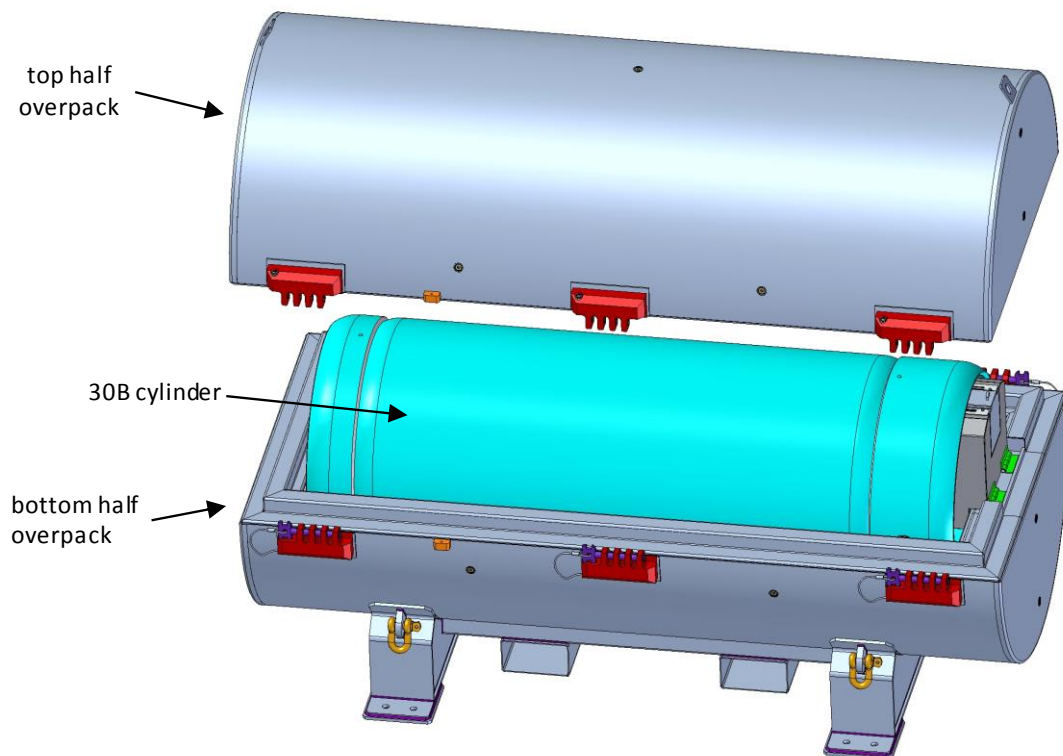


Figure 1: DN30 package overview

DESCRIPTION OF RADIOACTIVE CONTENTS

The DN30 package can be used for the transport of both commercial and reprocessed UF₆ with enrichments of up to 5 wt.% ²³⁵U, and may contain any mass of UF₆ from heels up to the maximal 30B cylinder fill limit of 2277 kg, which is equivalent to 1540 kg uranium.

The commercial grade UF₆ content is in compliance with ASTM C 996 [3] and may contain, in addition to the bulk of ²³⁵U and ²³⁸U, only small amounts of ²³⁴U and ²³⁶U as well as miniscule amounts of ²³²U (10⁻⁸ wt.-%) and ⁹⁹Tc (10⁻⁶ wt.-%).

Reprocessed UF₆ content is also based on [3], but there is an additional reprocessed UF₆ inventory option in the certificate for contents that exceed the values given there. In either case, the UF₆ may also contain impurities (fission products and higher actinides) and significantly larger amounts of the uranium isotopes ²³²U, ²³⁴U and ²³⁶U than for the composition defined in [3].

All UF₆ contents, whether commercial or reprocessed, contain very few neutron emitters, whose contribution to e.g. the external surface dose rate is < 2 μSv/h, so that the rest of this paper will focus on gamma emitters and dose rates.

SOURCE TERM CONSIDERATIONS

For commercial grade UF₆, the source term is dominated by the daughter products from uranium decay rather than the uranium isotopes themselves. The build-up of these decay products over time leads to an increase of the dose rates for a given UF₆ cylinder over time. This is reversed to the behaviour of most irradiated materials, and therefore a somewhat unusual situation for dose rate assessments.

The main contributions to the source term for a 30B cylinder with commercial grade UF₆ are from daughter nuclides of ²³⁸U, which makes up 95-100 % of the uranium content in the UF₆, and from the ²³²U daughter ²⁰⁸Tl. Given the miniscule concentration of ²³²U, the latter is surprising at first glance, but ²⁰⁸Tl produces extremely hard gamma radiation with an energy of 2.6 MeV, which can escape the UF₆ self-absorption relatively easily and will therefore cause high external dose rates.

For reprocessed UF₆, there are significant additional dose rate contributions from the fission products and higher actinides, but for ²³²U contents usually encountered in reprocessed uranium, the overall behaviour will be very straightforward: the gamma source term will be dominated by the contribution from the ²³²U daughter nuclide ²⁰⁸Tl.

The strong impact on external dose rates of both the initial ²³²U concentration and the build-up of ²⁰⁸Tl is reflected in the allowable concentration of ²³²U for reprocessed UF₆ contents: the maximum concentration is depending on the storage time between processing and the start of transport. For unlimited storage times, the ²⁰⁸Tl/²³²U equilibrium will be reached after approx. 10 years, and the source term will not increase after that point. The corresponding maximal ²³²U concentration is 0.02 μg per gram uranium (μg/gU). For transport within one month of processing, the limit for the ²³²U concentration increases to 0.06 μg/gU.

GEOMETRY MODEL FOR SHIELDING CALCULATIONS

A simplified geometry model of the DN30 package is used for shielding calculations: basically, only the steel shell of the 30B cylinder and steel shells and the PIR foam of the DN30 PSP are taken into account. The domed heads of the 30B cylinder are simplified to a flat shape, with the cylinder length extended up to the inner DN30 shell (see Figure 2). For full cylinders, the whole 30B volume is assumed to be filled with UF₆. With the UF₆ density of 4.68 g/cm³ assumed in the calculation – a covering low value to minimize self-absorption - this results in 3912 kg UF₆, still significantly more than the fill limit of 2277 kg. Since the source term is based on specific UF₆ activities, this assumption covers all possible UF₆ configurations for partial fillings.

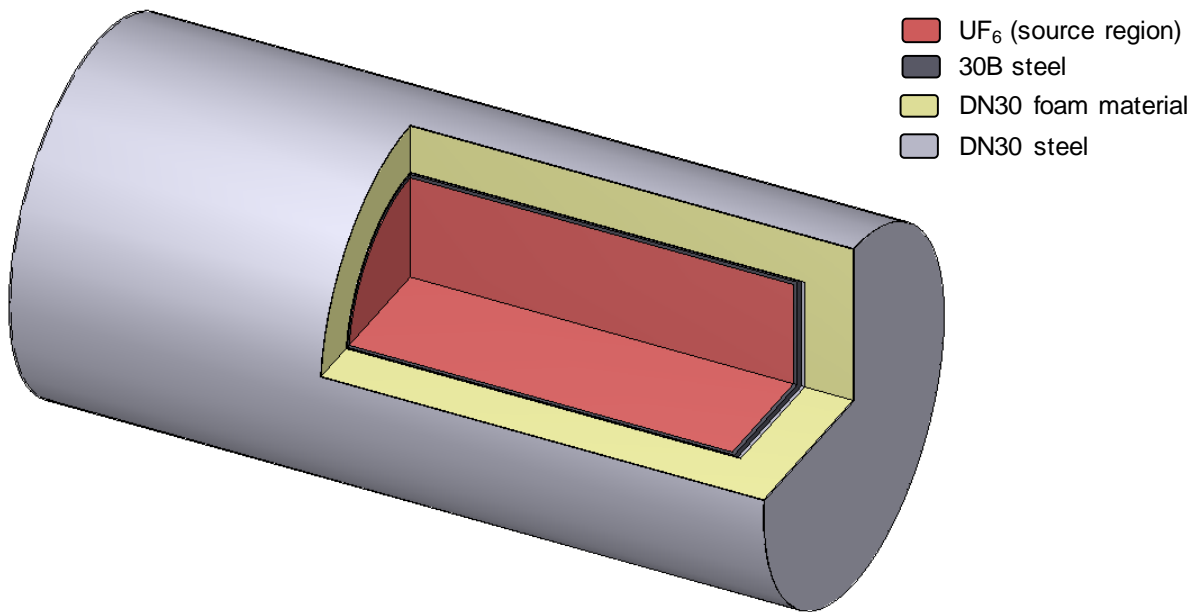


Figure 2: DN30 shielding calculation model with full UF₆ cylinders

For cylinders containing heels, two basic configurations of the radiation source (the UF₆ heels) were investigated: a thin layer of UF₆ attached to the inner side of the cylinder wall (model 1) or an accumulation of UF₆ in form of a puddle at the lower side of the cylinder (model 2, see Figure 3). Model 1 reflects the possibility that an inner layer of non-readily volatile uranium compounds adheres to the inner cylinder surface. Model 2 simply follows from the concentration of non-volatile compounds at the bottom, as the cylinders are emptied in horizontal position. The latter model leads to higher external dose rates both on the package surface and in 1 m or 2 m distance and is therefore used in the dose rate analysis.

An additional configuration with a thin layer attached to the inner side of the cylinder head (model 3, also in Figure 3) was also investigated in order to have a comprehensive analysis of the possible heels arrangements. This configuration, however, was not considered to be realistic enough to be taken into account for the dose rate analysis.

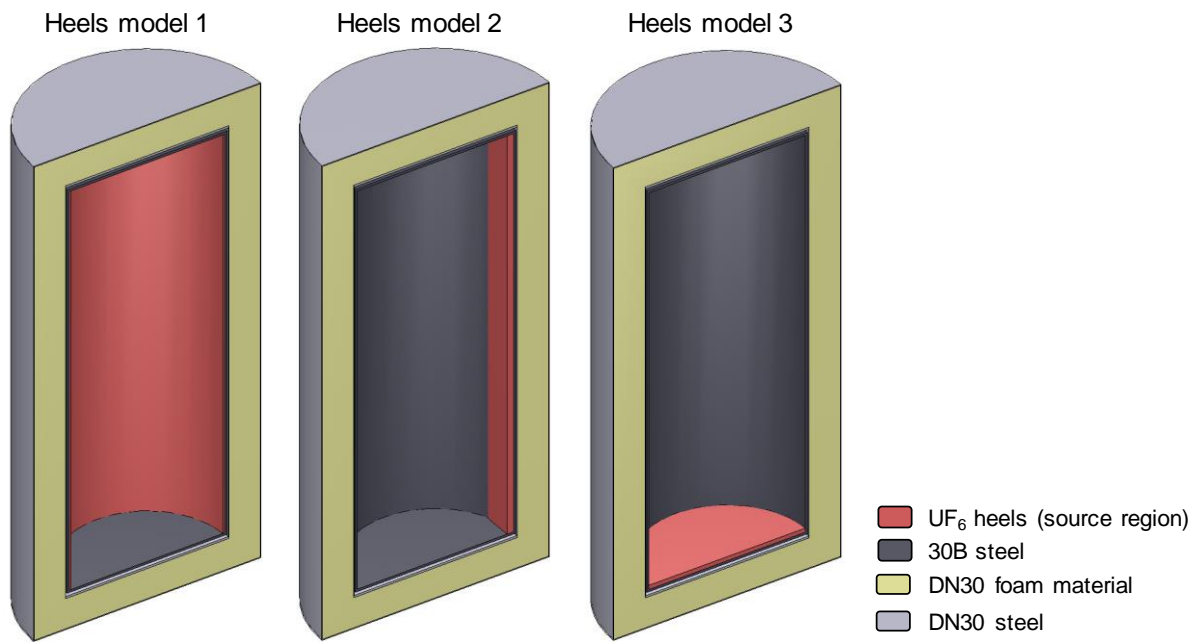


Figure 3: DN30 shielding calculation models with UF₆ heels cylinders

EXTERNAL DOSE RATES FOR FULL 30B CYLINDERS

With the assumptions given above, the calculated dose rates for the DN30 package with full cylinders are below the dose rate limits of the IAEA transport regulations (2 mSv/h at the package surface and 0.1 mSv/h at 2 m distance from the transport vehicle). The maximal dose rates occur in radial direction of the package. For the 2 m value, it is assumed that two packages are positioned face to face (see Figure 4), which results in the highest dose rate compared to other configurations of adjacent DN30 packages.

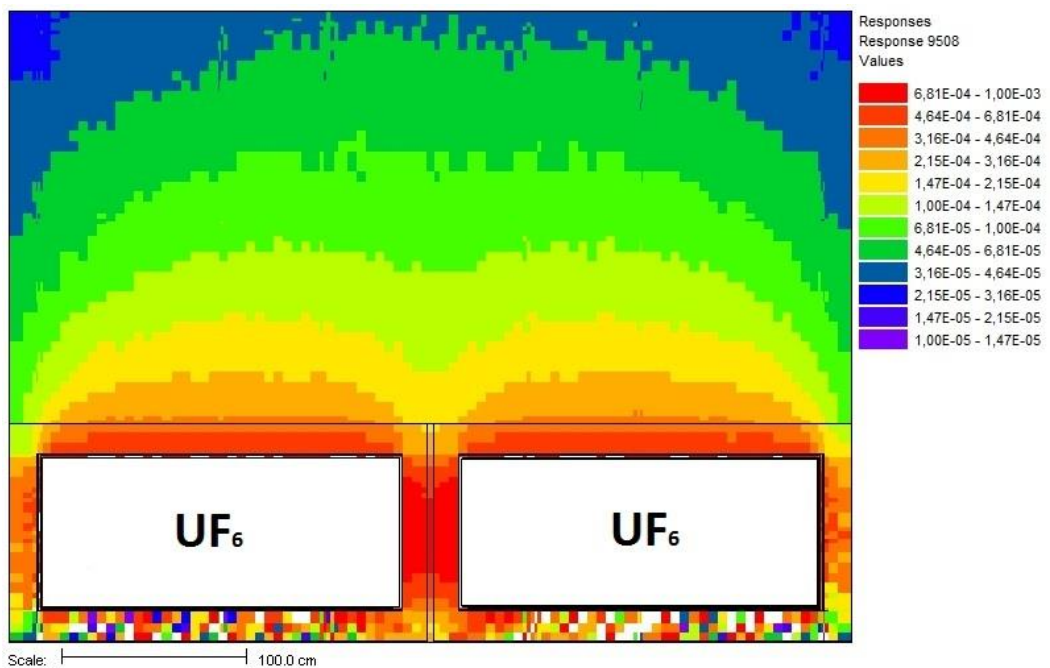


Figure 4: Dose rate distribution for two DN30 packages positioned face to face

The maximal surface dose rate for commercial grade UF₆ is 12 μSv/h, a very small amount. For reprocessed UF₆, it is 720 μSv/h, still less than half of the regulatory limit value. As noted above, the dose rate contribution from neutrons is very small (< 2 μSv/h).

Maximal dose rates at 2 m from the vehicle are 1 μSv/h for commercial grade UF₆ and 98 μSv/h for reprocessed UF₆, just below the regulatory limit of 100 μSv/h. The 2 m external dose rate is the main reason behind the linked restrictions on storage time before transport and on ²³²U content for reprocessed UF₆ contents.

SOURCE TERM DETERMINATION FOR UF₆ HEELS CONTENTS

Uranium heels are the remnants of UF₆ that may remain after emptying of cylinders. The maximum quantity of heels in a 30B cylinder in accordance with [1] is 11.4 kg UF₆. At first glance, this small UF₆ content should be covered by the dose rate analysis of a full 30B cylinder with 2277 kg UF₆, but the opposite is true: both calculated and measured external dose rates at cylinders containing heels can substantially exceed those of a full cylinder.

This is due to the behaviour of the impurities and uranium decay products that make the strongest contribution to gamma radiation: since these non-uranium nuclides will usually not form volatile compounds with fluoride, they are likely to remain in the cylinder when the UF₆ content is liquified during emptying. This leads to a concentration of the gamma emitters closer to the external surface for heels cylinders, which is reflected in the UF₆ heels configurations shown in Figure 3.

In the dose rate analysis for the DN30 package, it is assumed that all non-uranium nuclides from a full cylinder will remain in the heels quantity. These non-uranium nuclides also include decay products of uranium (most importantly, of the relatively long-lived ²³²U daughter ²²⁸Th, whose further decay leads to the strong gamma emitter ²⁰⁸Tl) that have built up during the time between processing and emptying of the cylinder.

Basically, a heels cylinder will have all the source intensity of a full cylinder, but concentrated close to the surface with almost no self-absorption in UF₆. While the assumption that 100% of the non-uranium nuclides will remain in the heels is an obviously conservative approach, it might not be too far away from the real behaviour of impurities in UF₆ when a cylinder is emptied.

It should be noted that unlike for a full cylinder, there will be no further dose rate increase with time, since further build-up of gamma emitters from the max. 11.4 kg of UF₆ heels is negligible compared to the decay of the remnant non-uranium nuclides from the full cylinder. Accordingly, the external dose rates at a heels cylinder will decrease with longer storage times after emptying.

For heels from commercial grade UF₆, there can be several instances of filling and emptying a cylinder without cleaning and washing in between. In the source term determination for the DN30 analysis, the decay products of uranium remain in the heels each time the cylinder is emptied and are added to the next filling of the cylinder. After undergoing a representative filling/emptying cycle, it is further assumed that the cylinder remains in filled condition for another 10 years to maximize the ²²⁸Th content resulting from the decay of ²³²U. Even with these pessimistic assumptions, the external dose rates are clearly below the regulatory limits (335 μSv/h at the package surface and 15 μSv/h in 2 m distance).

Heels from reprocessed UF₆ are more of a challenge, as explained in detail in the next section.

DOSE RATE ANALYSIS FOR HEELS FROM REPROCESSED UF₆

For heels from reprocessed UF₆, no refilling of cylinders is permitted without prior cleaning and washing, so the source term determination is based upon a single filling of the cylinder. As outlined above, the main source term contribution for typical compositions of reprocessed UF₆ will be from the ²³²U daughter ²²⁸Th, which decays further into the high-energy gamma emitter ²⁰⁸Tl.

This leads to a complex situation, where the external dose rate at a 30B cylinder with reprocessed UF₆ heels and at a DN30 package with such a cylinder depends on the following:

- initial ²³²U content (higher concentration → higher dose rate)
- build-up of ²²⁸Th between processing and emptying (longer times → higher dose rate)
- portion of non-uranium nuclides remaining in the heels (higher portion → higher dose rate)
- decay of ²²⁸Th between emptying and transport (longer times → lower dose rate)
- position of heels

For a typical reprocessed UF₆ content with an initial ²³²U concentration of 0.03 μg/gU, and a storage time between processing and emptying of half a year, the development of external dose rates over time after emptying is shown in Figure 5 (package surface) and Figure 6 (at 2 m distance). In these calculations, model 2 is used for the UF₆ heels geometry, and it is assumed that 100% of the non-uranium nuclides remain in the heels.

The calculation results clearly show the overwhelming contribution from the decay of ²²⁸Th, and how the dose rate decrease over time exactly follows the 1.9 years half-time of ²²⁸Th. For this UF₆ heels composition, the regulatory limits will be met after roughly 2 years for the surface dose rate and a bit more than a year for the 2 m dose rate.

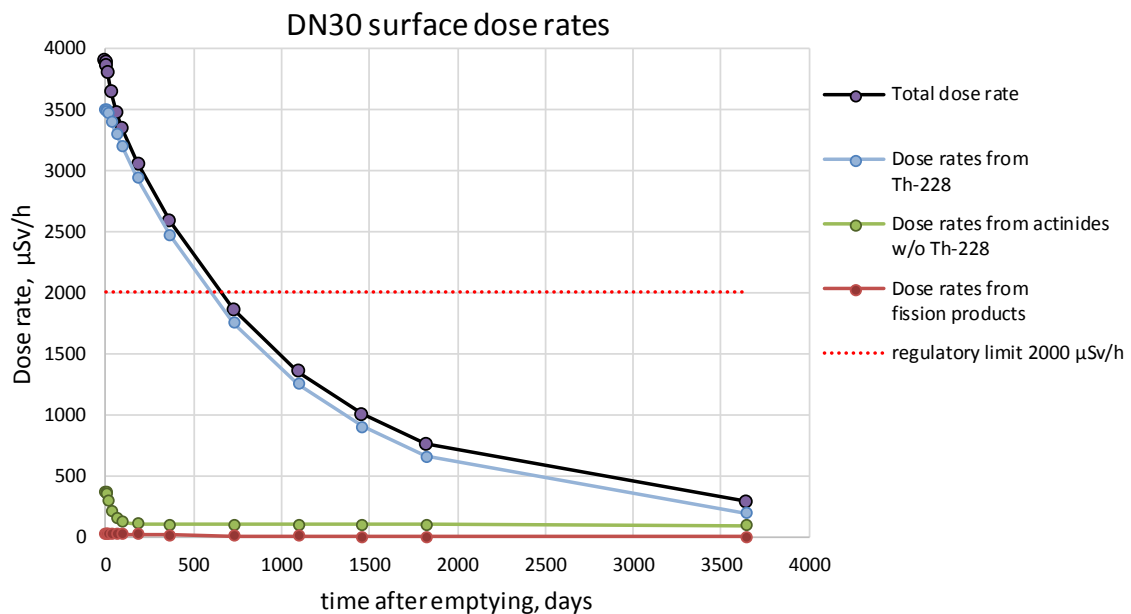


Figure 5: Surface dose rates for a DN30 package with UF₆ heels over time after emptying

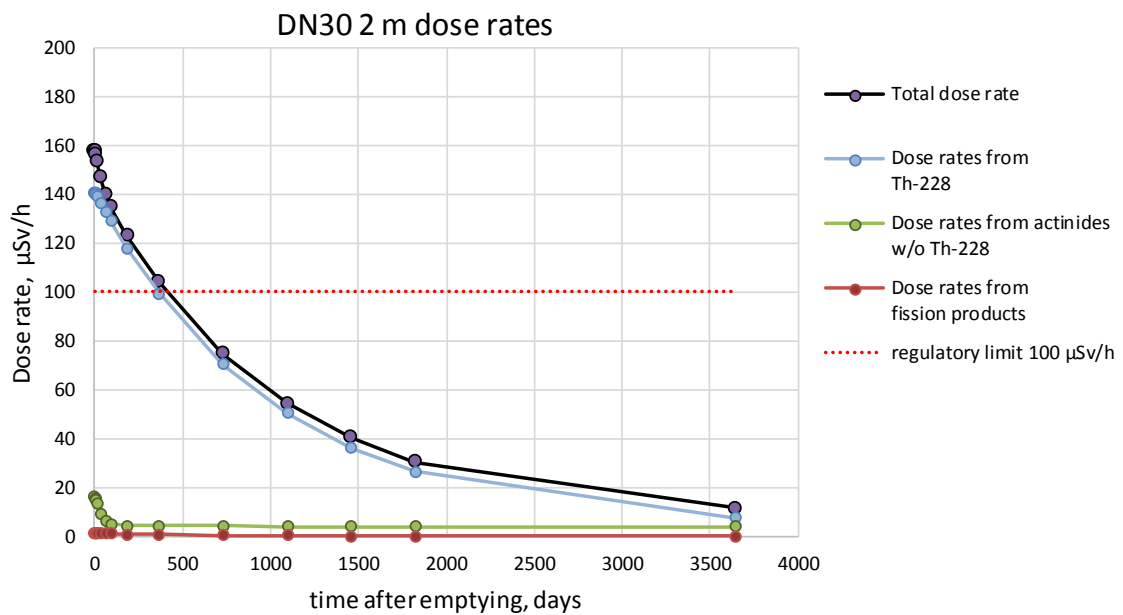


Figure 6: Dose rates at 2 m from a DN30 package with UF₆ heels over time after emptying

OPTIONS FOR CYLINDERS WITH EXCESSIVE DOSE RATES

Since the calculated dose rates shown above result from covering assumptions regarding the portion of non-uranium nuclides that remains in the UF₆ heels as well as the position of the heels in the 30B cylinder, real dose rates can be expected to lie below these calculation results.

To assess the transportability of real 30B cylinders in the DN30 package, the certificate provides the option to derive DN30 dose rates from measured dose rates at the bare 30B cylinder. Based on shielding calculations with all three heels geometry models shown in Figure 3, it was established that the maximum surface dose rate at a bare 30B cylinder is at least 2.5 times higher than at the surface of a DN30 package containing that cylinder. It follows that a bare 30B cylinder with a measured surface dose rate below 5000 μSv/h can be transported with the DN30 and meet the regulatory surface dose rate limit of 2000 μSv/h.

For reprocessed heels cylinders where the measured dose rate exceeds 5000 μSv/h, one option is to wait until the dose rate is low enough – as shown above, the decrease of the external dose rates follows the ²²⁸Th half-time of 1.9 years, so that their development over time can be accurately predicted.

If longer storage times after emptying incur too much additional costs or if storage place is limited, one option would be additional shielding on the DN30 package itself – the additional weight would be balanced by the decreased mass of the UF₆ content, since such a modified design would only be used for heels cylinders.

Depending on transport routes and means, another possibility would be to exploit the regulatory surface dose rate limit of 10 μSv/h for transports under exclusive use. In this case, only the external dose rate at 2 m from the vehicle would need to be reduced to below 100 μSv/h, which could be achieved by increasing the distance between the package and the outer extent of the vehicle, and/or additional shielding of the vehicle.

CONCLUSIONS

For the dose rate analysis of the DN30 package, a thorough investigation of the various contributions to the source term for 30B cylinders is used to derive the external dose rates via 3D shielding calculations. Dose rates for full cylinders with commercial grade or reprocessed UF₆ will generally comply with the regulatory limits. 30B cylinders with heels from reprocessed UF₆ with a high ²³²U concentration might exceed the regulatory dose rate limits for some time after emptying, but even for these cases, several options exist to allow an expeditious transport.

REFERENCES

- [1] ISO 7195, Nuclear Energy – Packaging of uranium hexafluoride (UF₆) for transport, Second edition, September 2005
- [2] Uranium Hexafluoride – Packaging for Transport, ANSI N14.1- 2012
- [3] ASTM C996, Standard Specification for Uranium Hexafluoride Enriched to Less Than 5 % ²³⁵U