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Criticality Analyses of Enriched Uranium-Hexafluoride Containing Impurities

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Abstract:

Due to its chemical and physical properties, enriched UF_6 presents several challenges to criticality safety. Recently, criticality safety evaluations of packages for the transport of enriched UF_6 have been increasingly subject to comprehensive examinations and verifications by the competent authorities worldwide.

In this paper, criticality safety of packages for the transport of enriched UF_6 (up to 5 wt.%) containing impurities is investigated. The investigation is based on variation calculations of criticality relevant parameters such as geometry and material composition for individual packages in isolation and arrays of packages. Single packages and infinite 3D arrays of packages were simulated to figure out most reactive arrangements and prove criticality safety for the DN30 overpack designed by DAHER-NCS.

Since UF_6 in 30B cylinders is under-moderated, the consideration of conservative amounts of moderation and the study of the impact of their geometrical distribution on reactivity are thereby of paramount importance. This involves the determination of all possible moderation sources resulting not only from inherent impurities in UF_6 (up to 0.5 wt.% HF) but also from the chemical interaction of uranium hexafluoride with possibly in-leaking water vapor.

Once these quantities of moderation have been determined, criticality analysis is performed for a variety of geometrical fuel-moderator distributions by means of conservative calculation models taking into account rigorous accident conditions of transport and effects related to the physical properties of UF_6.

The criticality safety was proven for the DN30 overpack, even though a very conservative, theoretical approach was taken in all assumptions and such hypothetical configurations may not be likely to be encountered in actual packaging, transportation, and storage configurations.

The high degree of similarity between UF_6 overpacks with regard to their geometry and manufacturing materials as well as the standardized use of 30B cylinders allows the applicability of the knowledge acquired in this analysis as well as the used methods to criticality safety assessments associated with other overpacks dedicated to the transport of enriched UF_6.

Introduction

The DN30 overpack was first presented during PATRAM 2010 [1] and is intended to be licensed as Type AF/IF/B(U)F for the transport of commercial grade and enriched reprocessed uranium with an enrichment of up to 5 wt.%. According to [2] and [3], subcriticality must be maintained during routine (RCT), normal (NCT) and accident conditions of transport (ACT). The proof of subcriticality involves not only the assessment of an individual package in isolation but also of package arrays. Since CSI=0, an infinite array of packages must be considered. The consideration of all sources of moderation represents therefore a challenge. This presentation is dedicated to the most reactive configuration under ACT.

Geometrical model

Based on both, the mechanical analyses and the results of the performed drop tests, any deformation or change in the geometry of the 30B cylinder can be excluded. It is therefore justified to model the 30B cylinder more realistically. The heads of the 30B cylinder are of a spherical shape and both skirts are taken into consideration as illustrated in Figure 1. The valve, plug and nameplate are however completely neglected. As to the dimensions of the 30B cylinder, the maximal dimensions specified in [4] are used.

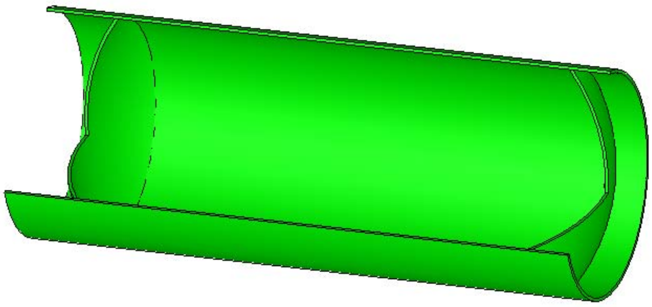


Figure 1: 30B cylinder

For RCT and NCT, an intact DN30 overpack has been considered, while an extremely damaged DN30 overpack is modeled for ACT. The shells of the damaged DN30 overpack are supposed to be compressed in such a way that no more foam is present and that its stainless steel sheets are pressed together to a single sheet in radial and axial directions as shown in Figure 2. This is a highly conservative assumption with regard to the mechanical analysis and the results of the drop test which has been briefly described in [5].

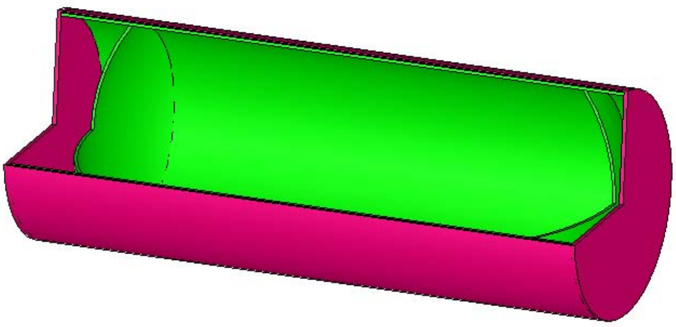


Figure 2: 30B cylinder accommodated in a damaged DN30 overpack (ACT)

Material compositions

Apart from structural materials, the quantitative determination of fissile materials and moderation sources is the first step toward a conservative assessment of the criticality safety of the DN30 overpack. Since the composition of UF_6 enriched to up to 5 wt. % U-235 is standardized in [6], the most conservative isotopic and chemical composition of UF_6 considered for the criticality safety assessments can be easily extracted and given below:

- Uranium enriched to 5.0 wt. % U-235
- It is conservatively assumed that 0.5 wt.% of the UF_6 is occupied by hydrogen fluoride (HF).

As to UF_6, a density of 5.5 g/cm³ has been used. This value is associated with a temperature of -40°C and has been derived from the diagram and data given in [7] by extrapolating beyond the densities given for higher temperatures.

According to [4], the fill limit for 30B cylinders is equal to 2277 kg which leads to a max. mass of 11,385 kg HF being present in a 30B cylinder. The density used for HF is equal to 1.15 g/cm³.

Beside inherent impurities, breakdown products have also a moderating effect and must be therefore determined. The chemical nature of UF_6 and particularly its aggressive behavior in the presence of water might result in a chemical interaction between UF_6 and possibly in-leaking water vapor during the long term use of 30B cylinders leading to the formation of certain amounts of breakdown products. The main challenge lies in the conservative determination of these amounts in which a complicated hydration chain results. The first reaction involving UF_6 in this chain is its hydration by water vapor which can be described as follows: $\mathbf{UF_6 + 2\,H_2O \rightarrow UO_2F_2 + 4HF}$

The main product of this hydration reaction is uranyl fluoride (UO_2F_2). The presence of even very small amounts of water or hydrogen fluoride triggers the hydration of uranyl fluoride which leads to the formation of uranyl fluoride hydrates. Uranyl fluoride hydrates could be present in many variations. However, a conservative reaction pattern resulting in the most hydrogenous breakdown products might be established as follows:

- Uranyl fluoride can be first hydrated by water vapor as per the reaction:

$$\mathbf{UO_2F_2 + x\,H_2O \rightarrow UO_2F_2{*}xH_2O} \qquad \mathbf{x_{max}=4}$$

- The uranyl fluoride hydrate can be subsequently hydrated by hydrogen fluoride as per the reaction

$$\mathbf{UO_2F_2{*}4H_2O + y\,HF \rightarrow UO_2F_2{*}4H_2O{*}yHF} \qquad \mathbf{y_{max}=3}$$

Due to the similar molar volume and molar weight of HF and H_2O and since hydrogen fluoride has half the moderation power of water, $UO_2F_2{*}4H_2O{*}3HF$ can be assimilated to $UO_2F_2{*}5.5H_2O$. Based on highly conservative assumptions related to the use of a 30B cylinder, up to 3.910 kg of $UO_2F_2{*}5.5H_2O$ might be present in a 30B cylinder at the end of a five years cycle.

Calculations and Results

A large number of variation calculations have been performed to figure out most reactive configurations. This includes most importantly the determination of an array structure leading to maximum effective neutron multiplication on the one hand, and on the other hand, the distribution of hydrogenous materials, 11.385 kg of HF along with 3.910 kg of breakdown products ($UO_2F_2{*}5.5H_2O$), in each package of a possible array.

Whenever the DN30 overpack is considered, only its steel shells are taken into account. In most cases however, only the bare 30B cylinder has been subject to calculations, which is an extremely conservative approach, considering the mechanical analysis and the results of the drop tests. Additional calculations have been performed to work out the minimum wall thickness of a 30B cylinder accommodated in the DN30 shells resulting in the criticality safety criterion being complied with.

The evaluation of the performed calculations indicates that hexagonal infinite arrays lead to higher k_{eff} than quadratic ones. Highest reactivity is calculated for an infinite hexagonal array composed of hexagonal prisms, in which the package (depending on model, only 30B cylinder or 30B cylinder accommodated in the compressed steel shells of the DN30 overpack) is surrounded by a water layer. The apothem of the hexagonal prism is such that it is equal to the outer radius of the package plus the thickness of the surrounding water layer. The rest volume of the hexagonal prism is kept empty (dry gussets) as schematically illustrated in Figure 3 for the case of bare 30B cylinders. UF_6 as material 500, carbon steel as material 2 and water as material 8 are plotted there in red, green and blue respectively.

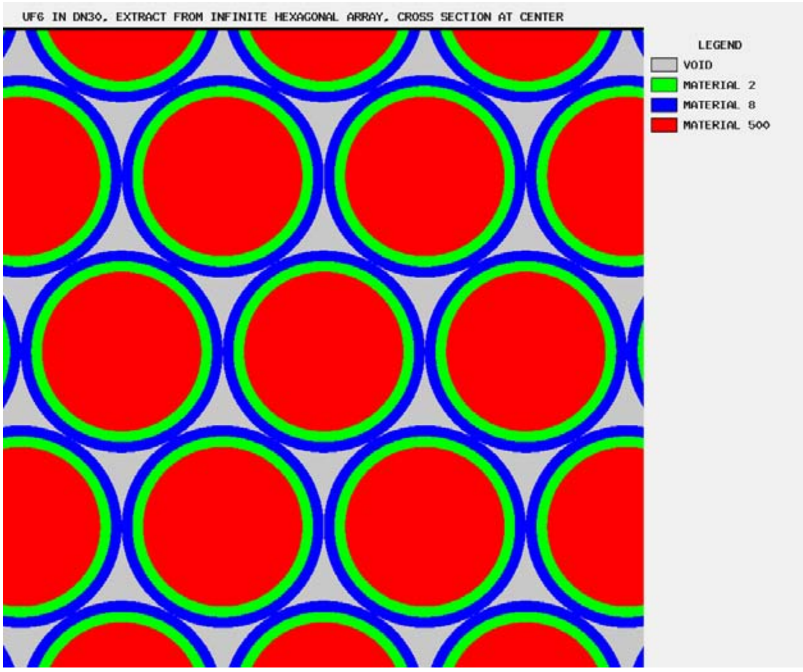


Figure 3: schematic illustration[1] of 30B cylinders in a hexagonal array

Numerous models have been considered with regard to the distribution of hydrogenous materials within the UF_6. In a first approach, HF and UO_2F_2*5.5H_2O have been homogenously distributed within the UF_6. However, such a distribution does not result in a high increase of k_{eff}. In a second approach, the hydrogenous materials have been concentrated into a lump surrounded by pure UF_6. The shape, size and the position of such a lump in the 30B cylinder affects significantly the reactivity of the system. For this reason, a variety of geometrical shapes has been treated. Above all, spherically shaped lumps such as a sphere and a spherical shell result in a significant increase of k_{eff}.

Since UO_2F_2*4H_2O*3HF, which has been assimilated to UO_2F_2*5.5H_2O, cannot be hydrated to a higher level by means of HF without becoming instable (y_{max}=3), further HF molecules such these associated with inherent impurities cannot be fixed in the UO_2F_2*4H_2O*yHF$|_{y=y_{max}}$ system. Thus, impurity lumps composed of separated breakdown products and inherent impurities have been also taken into account. Based on the two shapes leading to a high increase in reactivity, (sphere and spherical shell), an impurity arrangement composed of a sphere surrounded by a spherical shell has been considered.

Two cases are considered for this impurity arrangement. In the first case, an impurity sphere composed of 11.385 kg of HF is surrounded by a concentric spherical shell containing 3.910 kg of UO_2F_2*5.5H_2O. In the second case, the impurity sphere is supposed to be composed of UO_2F_2*5.5H_2O, whereas the surrounding concentric spherical shell is considered to contain HF.

The main task was to first determine the most reactive size of the impurity arrangement and subsequently to find out the position within the 30B cylinder leading to the highest k_{eff}. For this reason, the impurity arrangement has been modeled first at the centre of the 30B cylinder, which is referred to as the center of the UF_6. The outer radius of the concentric spherical shell has been varied, in order to figure out the most reactive impurity arrangement. The outer radius is varied in such a way that the total mass of the respective hydrogenous material is kept constant. Figure 4 shows the reactivity as a function of the outer radius of the concentric spherical shell for both cases. An impurity sphere composed of UO_2F_2*5.5H_2O being surrounded by a concentric spherical shell containing HF leads to higher k_{eff} values than for the opposite configuration. Furthermore, a HF spherical shell of an outer radius of 16 cm, and by implication an inner radius of 12.01 cm, as well as a UO_2F_2*5.5H_2O sphere of a radius of 8.51 cm, associated with a reflecting water layer of 0.8 cm leads to the highest reactivity. The volume between the sphere and the spherical shell is taken up by UF_6 as the rest volume of the 30B cylinder. This particular impurity arrangement is illustrated in Figure 5. HF as material 10 and UO_2F_2*5.5H_2O as material 503 are plotted there in pink and orange respectively.

[1] Thickness of cylinder side wall and reflecting water layer are increased for clarity reasons.

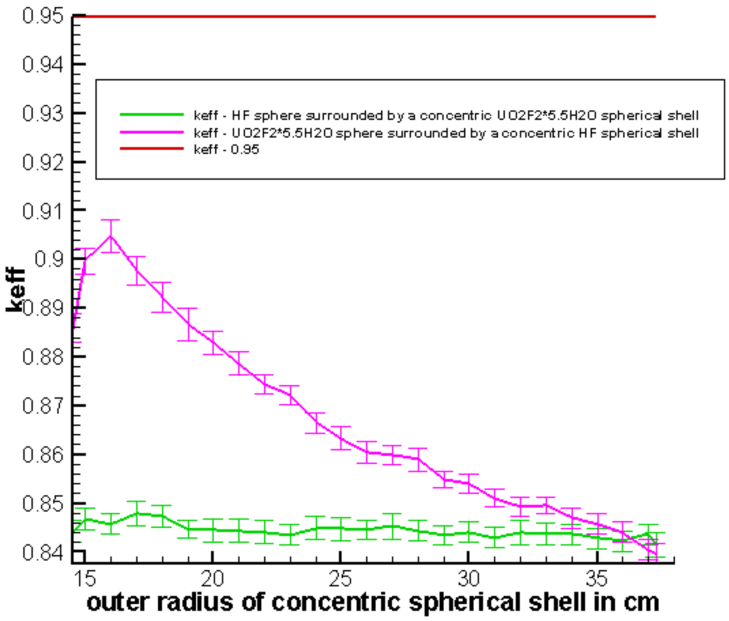


Figure 4: Reactivity as a function of the outer radius of the concentric impurity spherical shell for the centered impurity arrangement (error bars=3 σ)

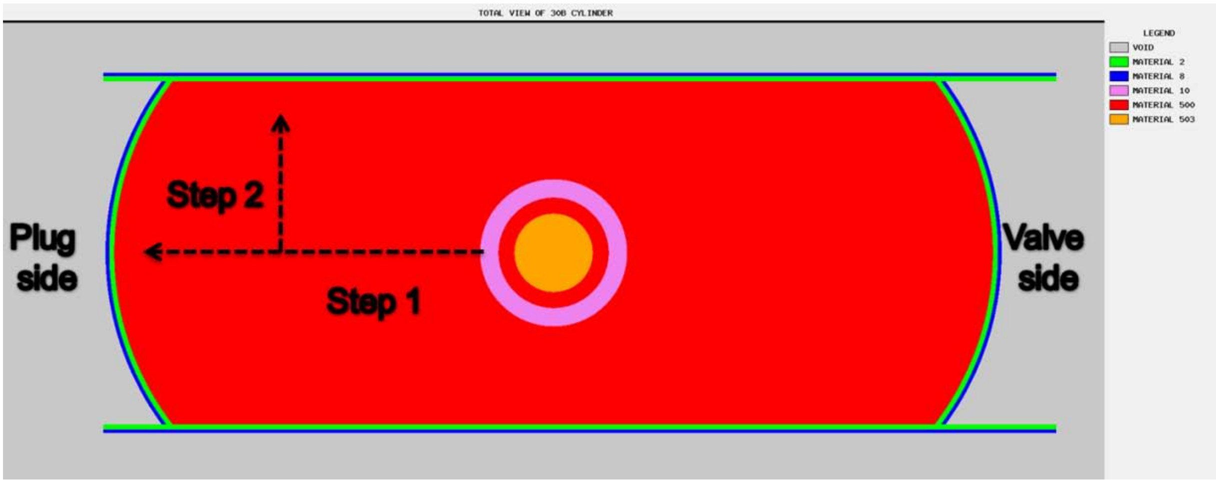


Figure 5: longitudinal section of a 30B cylinder containing the most reactive centered arrangement, outer and inner radius equal to 16 cm and 12.01 cm respectively

After having determined its most reactive size, the most reactive impurity arrangement is shifted toward the cylinder head associated with the plug side in a first step and toward the side wall of the 30B cylinder in a second step as indicated in Figure 5 to work out the position leading to the highest k_{eff} in the 30B cylinder. Here, the axial distance between the center of the 30B cylinder and the center of the impurity arrangement is varied until the head of the 30B cylinder and the impurity arrangement are in contact. For every considered axial distance between the centers, the radial distance is also varied until the side wall of the 30B cylinder and the impurity sphere are in contact. Furthermore, the thickness of the reflecting water layer around the 30B cylinder is varied for each single position. The variation of reactivity as a function of the position of the impurity arrangement within the 30B cylinder is shown in Figure 6. Two configurations are possible when moving the impurity arrangement toward the 30B cylinder side wall: the two- and three-arrangements-models as illustrated in Figure 7. However, the difference in reactivity is very slight and does not exceed 50 pcm for the worst case.

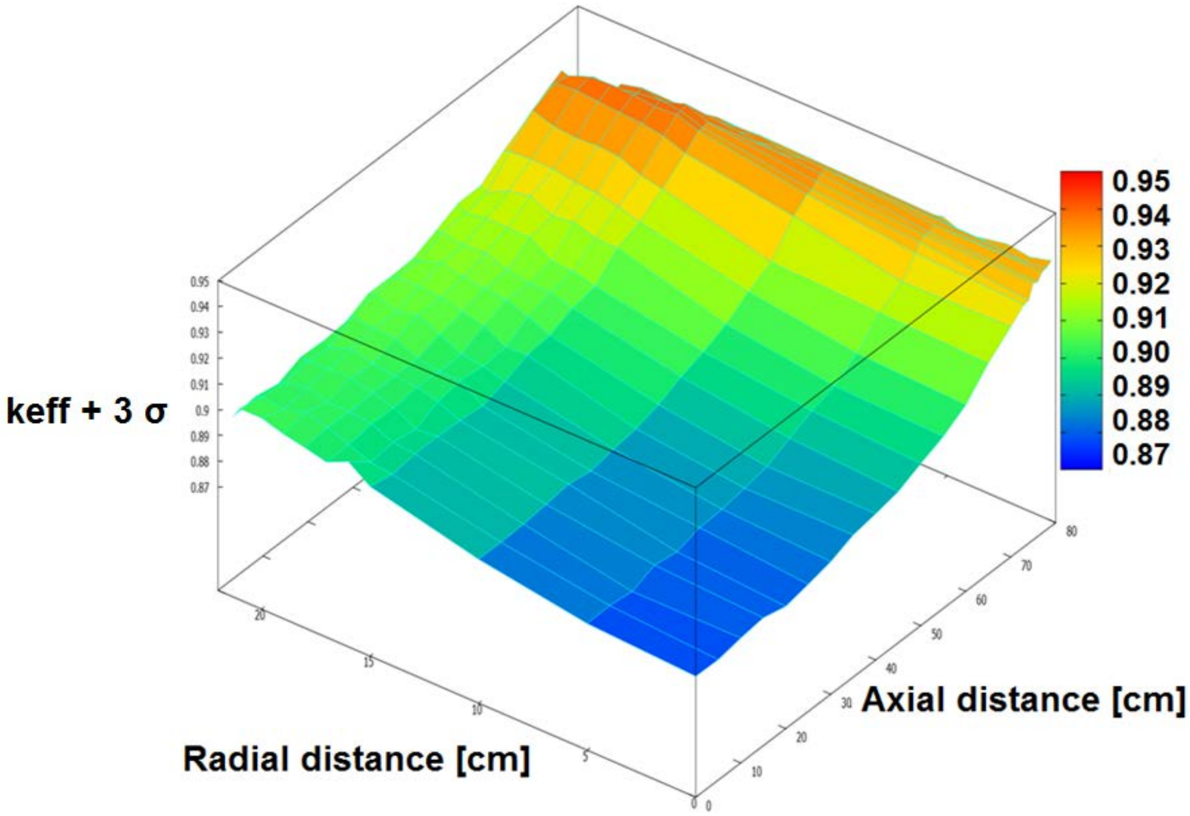
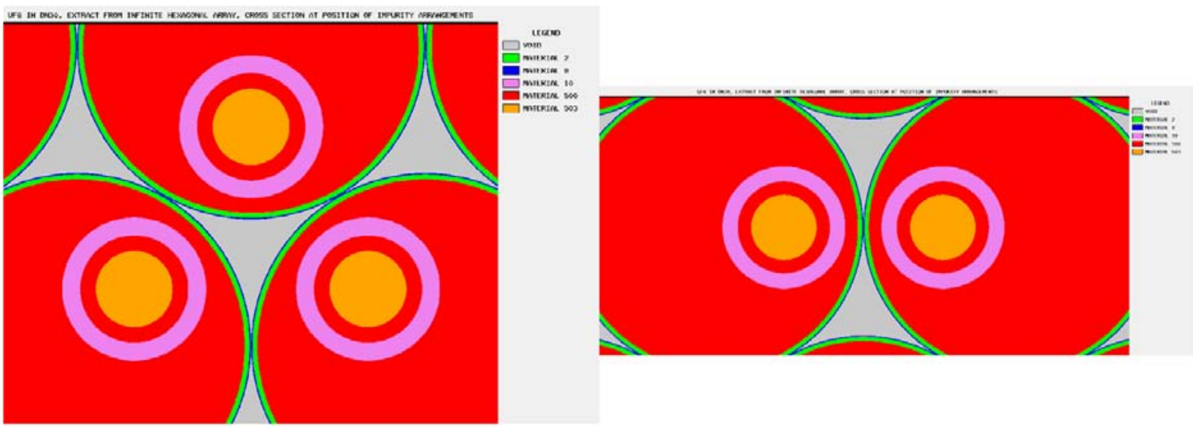


Figure 6: Reactivity as a function of the position of the impurity arrangement within the 30B cylinder, thickness of reflecting water layer is equal to 0.3 cm

The most reactive position of the impurity arrangement is located in immediate proximity to the 30B cylinder head and side wall. A radial and axial distance between the centers of the 30B cylinder and the impurity arrangement of 18 cm and 74 cm respectively associated with a reflecting water layer of 0.3 cm lead to the highest reactivity **k_{eff}+3σ=0.9428**. The criticality safety criterion is complied with for this highly conservative configuration.



Three-arrangements-model Two-arrangements-model

Figure 7: The two possible arrangements associated with the shifting of the impurity arrangement toward the 30B cylinder side wall

The impact of lower quantities of UF_6 has been also thoroughly investigated. For the most reactive configuration presented above, the amount of modeled UF_6 was diminished by considering lower UF_6 fill levels within the 30B cylinders. The axial fill level is varied from 40 cm, consistent with an almost empty cylinder, to 191.3 cm consistent with a completely full cylinder as shown in Figure 8. The variation of criticality as function of the UF_6 fill level is given graphically in Figure 9.

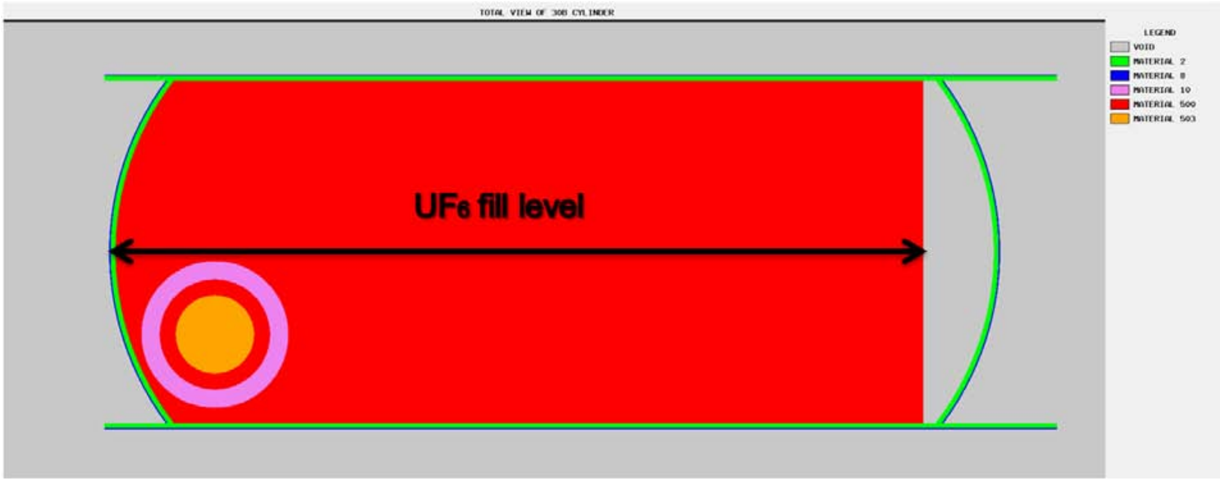


Figure 8: longitudinal section of a 30B cylinder containing the most reactive arrangement being at the most reactive position with a varied UF_6 fill level

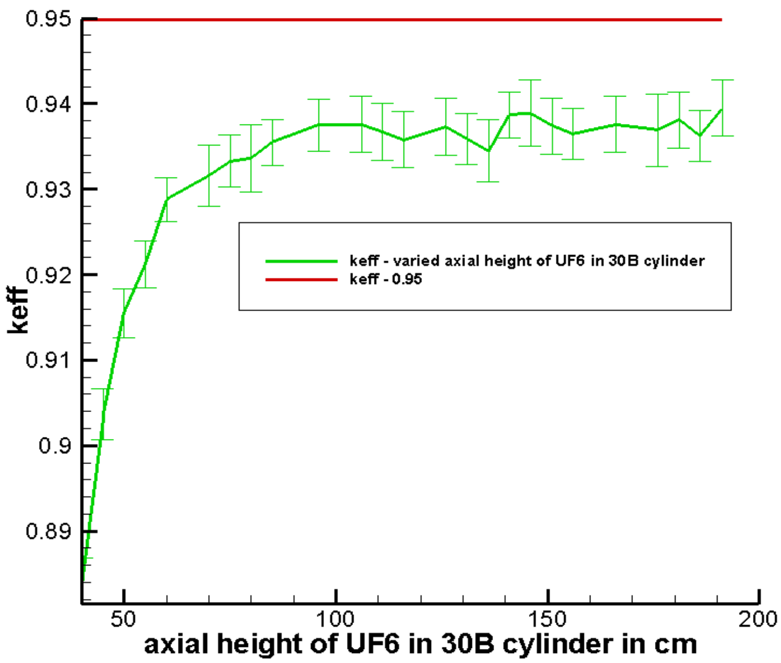


Figure 9: Reactivity as a function of the UF_6 fill level within the 30B cylinder (error bars=3 σ)

Figure 9 shows that the reactivity increases very steeply for small amounts of UF_6. For quantities equal or above the fill limit of the 30B cylinder (2277 kg), the reactivity fluctuates rather slightly around the reactivity reached for a completely filled 30 cylinder. In other words, the reduction of the considered quantity of UF_6 in the 30B cylinder does not have any increasing effect on the reactivity of the system.

A bare 30B cylinder has been treated so far. A wall thickness of at least 1.1 cm was necessary for the criticality safety criterion to be complied with. However, the consideration of the damaged DN30 overpack (compressed stainless steel shells) results in the criticality safety criterion being complied with even for thinner 30B cylinder walls. The evaluation of performed calculations has revealed that the criticality safety criterion is complied with for a wall thickness of 0.794 cm, consistent with the minimum wall thickness for a 30B cylinder as specified in [4], when taking the damaged DN30 overpack into consideration. The highest reactivity reached for this case was equal to **$k_{eff}+3\sigma=0.9237$**. As a consequence, there are no constraints regarding the wall thickness of the 30B cylinder if the damaged DN30 ovepack is taken into consideration.

Conclusions

The proof of criticality safety for the DN30 overpack has been a challenging task especially with regard to the infinite array and the relatively high amount of hydrogenous materials to be considered. The theoretical case of concern is associated with regular moisture ingress, which would cause UF_6 to react and form a hydrated uranyl fluoride (UO_2F_2). However, these deposits do not present a criticality hazard.

The most reactive scenario related to the impurity arrangement defined above is highly theoretical, because such a hypothetical configuration is not likely to be encountered in actual packaging, transportation, and storage configurations.

The criticality safety criterion is complied with even for bare 30B cylinders as long their wall thicknesses are greater than 1.1 cm. For 30B cylinders accommodated in DN30 overpacks, there are no criticality concerns with wall thicknesses greater than the min. permissible wall thickness of 0.794 cm as per [4].

References

[1] F. Hilbert, W. Bergmann, F. Noyon. PATRAM 2010: The DN30 overpack – A new solution for 30B cylinders

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[3] Regulations for the Safe Transport of Radioactive Material, 2012 Edition, SSR-6, IAEA, Vienna

[4] ISO 7195, Nuclear Energy – Packaging of uranium hexafluoride (UF_6) for transport

[5] W. Bergmann, T. Breuer PATRAM 2013: Drop testing of the DN30 protective structural packaging for the transport of Uranium-Hexafluoride

[6] ASTM C 996: Standard specification for Uranium Hexafluoride Enriched to less than 5% U-235

[7] USEC-651: The UF_6 Manual – Good Handling Practices for Uranium Hexafluoride, Revision 9, 2006