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REGULATORY CRITICALITY SAFETY REVIEW OF URANIUM HEXAFLUORIDE TRANSPORT PACKAGE APPLICATIONS

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ABSTRACT

Uranium hexafluoride (UF₆) has been safely transported across the world in 30B cylinders for many decades; however past history, although a good indication, does not guarantee future performance.

As a UK regulator for the transport of radioactive material, applications for UK transport package approval are received from all over the world. Judgement of their safety can only be made on the information provided in the safety case.

The criticality safety of a number of uranium hexafluoride transport applications have been recently reviewed. Assumptions have been made regarding the thickness of the 30B cylinder steel shell, the distribution of the hydrogen fluoride (HF) impurities and the potential for the uranium hexafluoride residues to hydrolyse. These assumptions each have an impact on the criticality safety of the package and will be discussed in turn.

From the results presented in this paper, it is considered that criticality safety cases which do not claim for the presence of the overpack will have difficulty in justifying that transporting uranium hexafluoride in these packages will be acceptable from a criticality safety perspective.

BACKGROUND

There are a number of packages that safely transport enriched uranium hexafluoride around the world. The most common is the UX-30 transport package that accounts for around 90% of the world market.



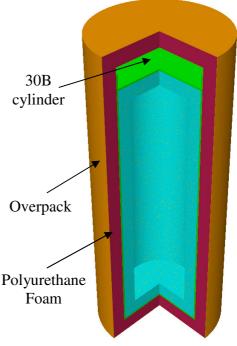


Figure 1. Uranium Hexafluoride Transport Package
- UX-30 (Reference 1)

Figure 2. Cutaway of UX-30 Transport Package

Although these packages use differing overpacks, the internal 30B cylinder used to contain the uranium hexafluoride is the same. These 30B cylinders are manufactured and maintained to the same ANSI or ISO standards (References 2 and 3 respectively).

CRITICALITY SAFETY

These uranium hexafluoride transport packages can contain up to 2277kg of 5.0wt% U-235/U enriched fissile material so the demonstration of criticality safety is of paramount importance.

The criticality safety case for many of these packages do not claim credit for the overpack. Although a significant pessimism, this allows the criticality safety report to be used in a variety of safety cases that have differing overpacks. Consequently, criticality safety reports such as the well respected Oak Ridge National Laboratory report presented in Reference 4 have supported the safety case for uranium hexafluoride transport packages for a number of decades.

Corrosion of 30B Cylinder Steel Shell

However, the 30B cylinders used to transport uranium hexafluoride can be in service for many decades. The environmental conditions that these cylinders are exposed to and the caustic nature of the contents can lead to corrosion of the steel of the 30B cylinder.

This is a recognised effect and an allowance for this is made in the ANSI and ISO standards such that 30B cylinders should be removed from service when their shell thickness has decreased to below $\frac{5}{16}$ " (0.794cm) from a nominal thickness of $\frac{1}{2}$ " (1.270cm).

The information presented in References 5 and 6 indicates that depleted uranium hexafluoride storage cylinders made from mild steel can experience external corrosion that gradually and uniformly reduces the thickness of the metal over time. It is stated that the external corrosion rate for these types of cylinders could be up to 0.05mm per year. If the same rate of corrosion is assumed to occur for 30B cylinders, this equates to a loss in steel mass of up 2kg per year. The internal corrosion rate for cylinders carrying enriched uranium hexafluoride is currently undetermined.

The current UK validation certificate for the UX-30 transport package, USA/9196/AF-96, limits the mass of uranium hexafluoride carried from 455kg to 2277kg. This is due to the possibility that following a fire, the internal pressure could exceed appropriate maximum limits when less than 455kg of uranium hexafluoride is carried. However, as not all of the uranium hexafluoride will be removed following the emptying of the 30B cylinder, a residue mass limit of up to 12kg is allowed.

Compliance with Uranium Hexafluoride Residue Mass Limit

In order to demonstrate compliance with the residue mass limit of 12kg, the operators usually weigh the 30B cylinder with the baseline tare weight being taken when the cylinder is either new or after it is washed out. However, as it can be up to five years between cylinder washes, a baseline tare weight can potentially be up to five years out of date.

If corrosion has occurred to the 30B cylinder, then assuming the worst-case loss in steel mass of 2kg per year (as indicated above), this could lead to an overall loss in steel of up to 10kg over five years. If this is not taken into account, then the mass of residue calculated to be present by the tare weight measurement could be significantly underestimated.

Shell Thickness of 30B Cylinder

In criticality safety assessments, it is common practice to pessimistically model parameters so that the k_{eff} for the system is maximised. This means that for the 30B cylinder, instead of modelling a steel shell thickness of ½" (nominal), the $^{5}/_{16}$ " (minimum) should be modelled.

Figure 3 presents the limiting model in the criticality safety assessment – an infinite array of 30B cylinders.

It can be seen from this figure that the uranium hexafluoride is modelled as being along the internal shell walls of the 30B cylinder. This is a possible geometry if, as suggested in Section 2.4 of Reference 4, the 30B cylinder contains uranium hexafluoride in liquid form which then 'cools uniformly from the outside with a corresponding decrease in volume'. Thus, 'the resulting solid UF₆ typically has a void in the center'.

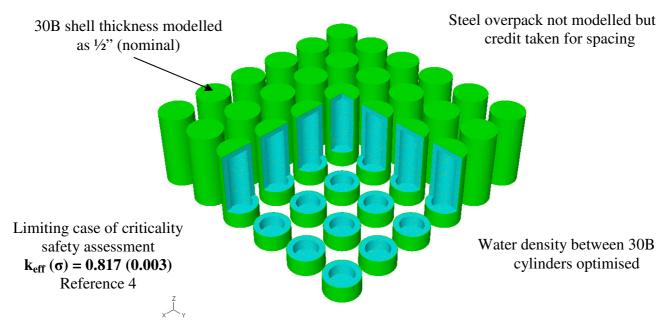


Figure 3. Infinite Array of 30B Cylinders

The limiting value of k_{eff} (σ) for the above model is 0.817 (0.003). However, the thickness of the 30B cylinder shell has been modelled as ½" (nominal). The model presented in Figure 4 has modelled the 30B cylinder shell thickness as $^5/_{16}$ " (minimum).

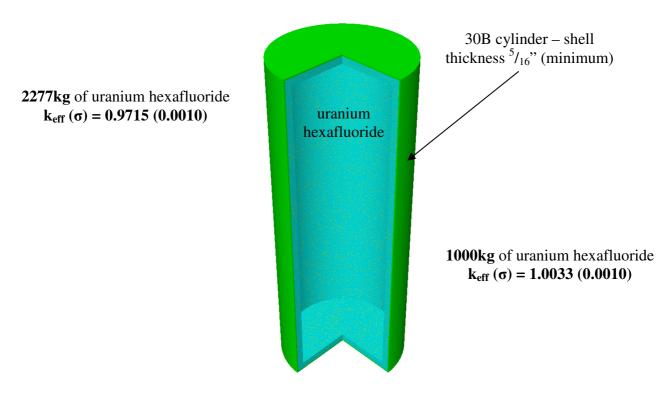


Figure 4. 30B Cylinders - Minimum Thickness of 30B Shell (5/16")

It can be seen from Figure 4 that reducing the thickness of the 30B cylinder shell from $\frac{1}{2}$ " (nominal) to $\frac{5}{16}$ " (minimum) increases the k_{eff} of the system by about 15% to over 0.97. This k_{eff} result now exceeds the criticality safety criterion of $k_{eff} + 3\sigma \le 0.95$.

However, it can be seen from the above figure that reducing the mass of uranium hexafluoride from the maximum allowed of 2277kg to 1000kg can lead to an increase in the k_{eff} of the system by about 3% so that the limiting k_{eff} now exceeds 1.0. Criticality safety has therefore not been demonstrated when only bare 30B cylinders are modelled.

From the results presented above, it is considered that criticality safety cases which do not claim credit for the presence of the steel overpack will have difficulty in justifying that transporting uranium hexafluoride in these packages will be acceptable from a criticality safety perspective; unless procedures are in place to ensure that the thickness of the 30B cylinder shell wall will not reduce below a certain acceptable minimum value.

Of course modelling the steel overpack leads to a large reduction in the k_{eff} of the system (~18%) as can be seen in Figure 5.

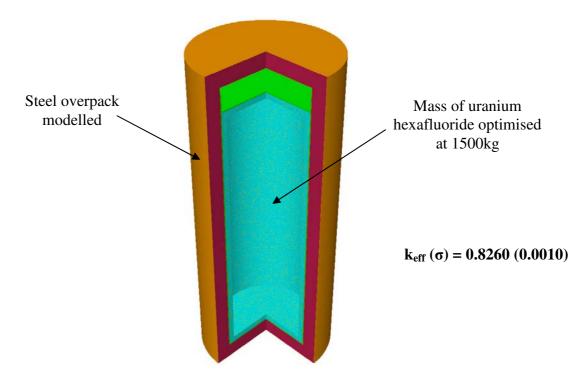


Figure 5. Overpack Modelled

Concentration of Hydrogen Fluoride Impurities

The fissile material carried within the 30B cylinder is not 100wt% pure uranium hexafluoride. An allowance is made for impurities of up to 0.5wt% which could be in the form of hydrogen fluoride.

The hydrogen fluoride is initially assumed to be homogeneously mixed with the uranium hexafluoride. In reality, this is not likely to occur as the density of hydrogen fluoride is about 1.0g/cc, lower than the density of uranium hexafluoride (in the liquid

phase) of about 3.0g/cc (Reference 7) rising to about 5.1g/cm³ when in the solid phase. The hydrogen fluoride will therefore rise through the uranium hexafluoride.

In the absence of any information regarding possible geometries that the hydrogen fluoride impurity could form, models were created in order to determine the worst-case system. A number of geometries were considered but the limiting case seemed to occur when the hydrogen fluoride was concentrated at full density in a spherical shell surrounding an optimum volume of uranium hexafluoride (modelled at a reduced density). This model is presented in Figure 6.

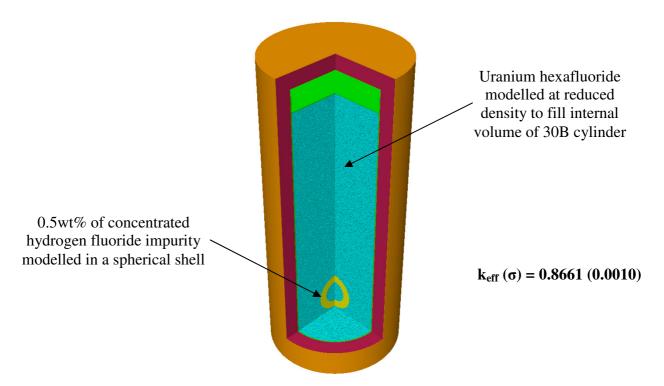


Figure 6. Concentration of Hydrogen Fluoride Impurities

An investigation into the neutronics of this model indicates that the hydrogen fluoride is acting as both a moderator and reflector of neutrons back into the centre of the optimised region of the low-density uranium hexafluoride. It can also be seen that this model achieves a k_{eff} (σ) for the system of 0.8661 (0.0010), an increase from the model presented in Figure 5 of about 4%.

In the absence of evidence to justify a homogeneous distribution of hydrogen fluoride throughout the uranium hexafluoride, it is believed that models such as that presented in Figure 5 above should be considered.

Hydrolysis of Residues

IRSN (Institut de Radioprotection et de Sûreté Nucléaire) have highlighted an issue whereby ingress of air (moisture) into the 30B cylinder could possibly occur during the engagement / disengagement of the filling apparatus as well as during long term storage and could, over time, lead to the hydrolysis of the uranium hexafluoride residues to produce uranyl fluoride (UO_2F_2). The equation for this reaction is:

$$UF_6 + 2H_20 --> UO_2F_2 + 4HF$$

Under worst-case conditions, the uranyl fluoride could hydrate to produce UO_2F_2 -5.5 H_2O with an H/U ratio up to 11.

It was seen above that the mass of residue allowed in the UK validation certificate for the UX-30 transport package is up to 12kg. The maximum allowed residue mass was assumed to hydrate to an H/U ratio of 11 and was located in the most reactive part of the model presented in Figure 6 (within the spherical shell of hydrogen fluoride). The density of the uranium hexafluoride is optimally modelled at full density. This model is presented in Figure 7.

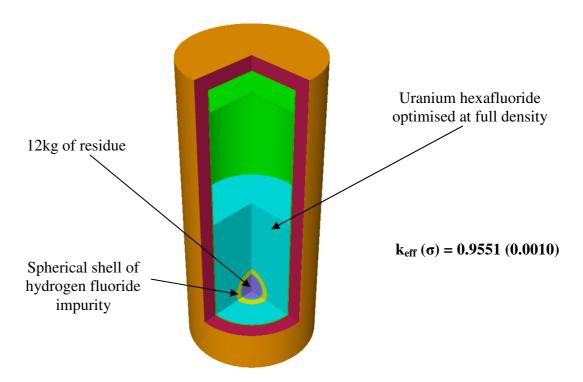


Figure 7. Hydrolysis of Uranium Hexafluoride Residues

It can be seen from the k_{eff} achieved for the model presented in the figure above that again the criticality safety criterion for the system, $k_{eff} + 3\sigma \le 0.95$, is exceeded albeit marginally. However, there are a number of pessimisms in the model that can be reduced; such as a finite array could be modelled, a lower residue mass limit could be assumed or a lower H/U hydration level of the residues could be justified.

CONCLUSION

Packages containing enriched uranium hexafluoride have been safely transport around the world for many decades. Some of the criticality safety cases used in support of these transports have been in place for many years and are well respected. Although some of the assumptions made in these safety cases are extremely pessimistic, some are not.

It is the view of the UK competent authority that the criticality safety cases used in the support of the transport of enriched uranium hexafluoride are reviewed to ensure that:

- Minimum 30B cylinder shell thicknesses have been used,
- Justification is provided for the hydrogen fluoride impurity distribution assumed and
- Appropriate hydration (H/U ratio) of the residues is considered.

From the results presented in this paper, it is considered that criticality safety cases which do not claim for the presence of the overpack will have difficulty in justifying that transporting uranium hexafluoride in these packages will be acceptable from a criticality safety perspective.

In addition, if the potential corrosion of the 30B cylinders is not taken into account, the mass of the residue left within the cylinder following discharge of the uranium hexafluoride could be underestimated.

REFERENCES

- 1. Source for Figure 1 www.topworkplaces.com/frontend.php/regional-list/company/news-record/columbiana-hi-tech
- 2. ANSI N14.1-2012, "American National Standard, For Nuclear Materials Uranium Hexafluoride Packaging for Transport", American National Standards Institute, Inc., Approved December 3, 2012.
- 3. International Standard ISO 7195:2005, Nuclear Energy Packaging of Uranium Hexafluoride (UF₆) for Transport.
- 4. ORNL/TM-I 1947, "Criticality Safety Review of 2½, 10 and 14 Ton UF₆ Cylinders", B L Broadhead, Martin Marietta Energy Systems, Oak Ridge National Laboratory, October 1991.
- 5. Portsmouth Gaseous Diffusion Plant Report POEF-323-94-42 "Evaluation of Ultrsonic Thickness Measurements of UF₆ Tails Cylinders", M. Lykins and A. Edler, Lockheed Martin Utility Services, Inc., December 1994.
- 6. Portsmouth Gaseous Diffusion Plant Report POEF-TS-02 "Review of Corrosion in 10- and 14-Ton Mild Steel Depleted UF₆ Storage Cylinders", Michael L. Lykins, Lockheed Martin Utility Services, Inc., August 1995.
- 7. Figure 5.2 of ORNL/ENG/TM-51, "Correlation of the Thermophysical Properties of Uranium Hexafluoride Over a Wide Range of Temperature and Pressure", J C Anderson, C P Kerr, W R Williams, Martin Marietta Energy Systems, Oak Ridge National Laboratory, August 1994.