UF_6 RELEASE CALCULATIONS AND RADIOLOGICAL AND ENVIRONMENTAL IMPACTS OF A UF_6 CONTAINER SUBJECT TO A LONG DURATION FIRE

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ABSTRACT

Assuming that a 12 tonne-UF_6 cylinder without thermal protection is subject to a long duration fire without intervention of the emergency response team, we have used the TENERIFE, PEECHEUR experimental results and the DIBONA model to evaluate the quantity of UF_6 that might be released and the associated chemical, radiological and environmental impacts.

The PEECHEUR experimental program, relative to the mechanical behaviour of a 48Y container subject to fire, has provided the bursting pressure and the size of the resulting break in container steel wall.

The DIBONA model, based on results of the TENERIFE experimental program relative to the thermodynamical behaviour of a UF_6 container in fire, has been used to evaluate the state of an unprotected 48Y container subject to an engulfing 800°C fire when the bursting pressure is reached.

Considering, at bursting time, the initial temperature distribution in the container and the UF_6 solid, liquid and gas phases repartition, the quantity of UF_6 gas that might be dispersed in the atmosphere has been calculated. Three steps are considered, the release of compressed UF_6 gas at bursting, then the evaporation of UF_6 liquid inside container, then the sublimation of the remaining UF_6 solid. Flow rates decrease from 300 kg/s down to 0,5 kg/s.

48Y containers containing 12 500 kg of natural UF_6 and 30B containers containing 2300 kg of enriched UF_6 are considered.

By assuming instantaneous hydrolysis of the released UF_6 from the container, atmospheric dispersion of HF and UO_2F_2 is calculated for several atmospheric conditions.

The chemical toxic consequences of exposition to UF_6 and UO_2F_2, depending on the distance from the release point, are calculated. The radiological consequences of exposition to uranium is only considered for UO_2F_2 enriched up to 5% of U_{235}.

For all configurations, it appears that:

- chemical risk due to HF is higher than the one due to UO_2F_2,
- concerning UO_2F_2, radiological risk is less important than chemical risk for low enriched, not reprocessed uranium.

Finally, the maximum distance from the release point where irreversible effects for population health are reached is about 5 kilometers (3 miles).

INTRODUCTION

UF_6 transportation packages exposed to long duration fire present risks of rupture of the containment system and of massive release of UF_6 into the atmosphere. Due to the imprecision of the calculation codes, these risks appear in the case of a 800°C fire with an approximate duration of 30 minutes or more. The objective of the study is to quantify the radiological and toxic consequences of such a release of UF_6. Package types 48Y and 30B are considered. They contain respectively 12 500 kg of natural UF_6 and 2300 kg of enriched UF_6. It is assumed that the fire is long enough to lead to the rupture of the container and conservatively we consider that the fire continues during the phase of release of UF_6.

After having determined the leakage flows, the consequences of the release of the UF_6 are calculated. It is considered that the released UF_6 has undergone a total and instantaneous hydrolysis to be transformed into HF and UO_2F_2. The analysis will notably enable the chemical effects of the toxicity related to the HF and the radiological effects related to the UO_2F_2 to be compared and to give the distance beyond which the release might have irreversible consequences for the population.

CONDITION OF THE CONTAINER PRIOR TO BURSTING

The container studied is a 48Y container containing 12 500 kg of UF_6 submitted to an engulfing fire at 800°C. The condition of the container, that is, the temperatures of the walls and the UF_6, the distribution between solid, liquid and gaseous phases as well as the internal pressure is determined from the results of the TENERIFE and PEECHEUR study programmes.

TENERIFE is a Franco-Japanese experimental programme carried out in co-operation with the IPSN and CRIEPI with the support of the EEC and French industrials [1] to [6]. The programme consisted in studying the behaviour of a container filled with UF_6 placed inside a furnace. On the basis of these tests, a thermodynamic model called DIBONA was developed [1] to [4]. The numeric use of this model enabled the evolution of the behaviour of a 48Y container exposed to a fire to be forecast and therefore to determine the condition of the container prior to bursting.

With a concern for simplifying the release calculation model, the temperatures are considered as homogeneous in liquid, in solid, at the liquid/gas interface and on the wall zones of the container in contact with the liquid and in contact with the gas. The temperatures are referred to in figure 1.

According to the results obtained using the DIBONA model [4], it was considered that prior to rupture of the container the solid represented 1/3 of the mass of the liquid UF_6 which gives the initial distribution detailed in figure 1:

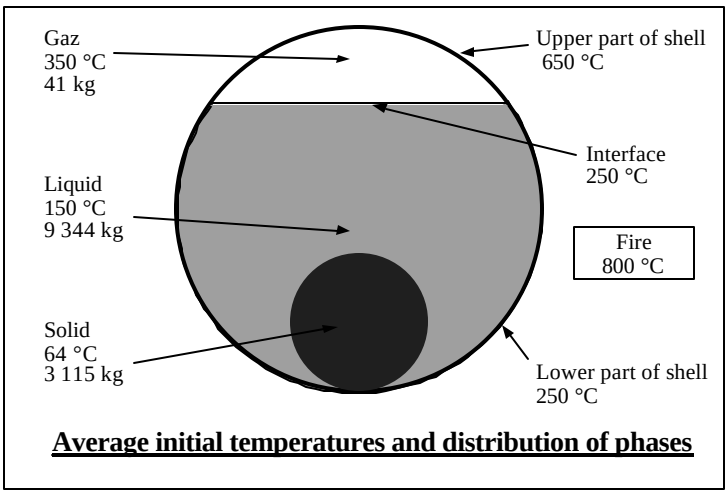

Figure 1: Condition of the 48Y container prior to bursting

PEECHEUR is an experimental programme led by the IPSN with support from COGEMA. Its objective was to study the rupture modes of a 48Y package submitted to internal pressure and to temperatures comparable to those determined using the DIBONA programme for a package submitted to a fire. This programme enabled to observe so that the rupture of the container takes place at pressures ranging from 40 to 52 bar [3], [6]. It is assumed in the follow up of the study that the rupture occurs for an internal pressure of **46 bar** which corresponds to the value currently admitted for the critical pressure of UF_6.

In addition, the results of the PEECHEUR programme [3], [6] allowed the breach consecutive to the bursting of the container to be modelled by a rectangular slit of dimension **200x55 mm²**.

CALCULATION OF RELEASE FLOW

Two configurations

Location of the breach depends of complex phenomena: the presence of metallurgical faults, homogeneity of the fire, position of the container in the fire, etc … .

To calculate the release flow of UF_6, let us consider two configurations depending on the location of the breach. The first configuration where the breach is below the free surface of the liquid is less likely since located in a relatively cold steel zone; however, we have chosen to consider that the breach is at the bottom point of the container thus allowing the complete drainage of the container. In the second configuration, let us consider that the breach is located above the free surface of the liquid. This configuration is more likely since the breach is located in a hotter and thus weaker steel zone.

Breach located in liquid phase

In the configuration where the breach is below the free surface, the UF_6 liquid is leaking spontaneously.

When considering that the flow of UF_6 in liquid phase is governed by a law of pressure loss at the breach, it is calculated that 26.5 seconds are needed to evacuate the 9 344 kg of liquid UF_6. This relatively short duration justifies that the spontaneous vaporization flow of UF_6 liquid at 12 bar, saturated vapour pressure of the liquid at 250°C, does not change the duration of drainage time.

Three successive phases characterise the behaviour of the liquid UF_6 coming out of the container:

- the UF_6 vaporizes when getting the energy needed for transformation in the cooling of the fraction of remaining liquid UF_6,
- the remaining UF_6 liquid reaching the triple point temperature (64°C), it continues to evaporate in getting the energy needed in solidification of the fraction of remaining liquid UF_6,
- the remaining solid UF_6 has been cooling since 64°C to 56.4°C, the temperature of the point of sublimation at atmospheric pressure, in getting the energy needed in sublimation of the fraction of solid. The solid UF_6 at 56.4°C may then continue to sublimate.

The experience resulting from the observation of accidental release of UF_6 has enabled to consider that the first two phases of vaporization and sublimation are instantaneous.

When considering that there is no contribution of energy to the liquid UF_6, energetic analysis shows that the UF_6 vaporized during the first two phases represents 68% of the liquid UF_6, that is, 6 354 kg. The remaining 32% (2 990 kg) have been solidified.

These 2 990 kg of solid sublimate outside of the container under the combined effects of the energy produced by the surrounding fire, the convective movements that remove the UF_6 vapours from the surface and the hydrolysis reaction of the UF_6 that promotes disappearance of the solid.

The kinetics of the sublimation is unknown, it is assumed however that the sublimation is rapid and that the 2 990 kg of solid UF_6 sublimates within the same lapse of time as the time taken for the liquid to vacate the container.

The remaining solid within the container following drainage is assumed to be at 64°C. The solid is overheated, thus 2.7% (86 kg) of UF_6 will spontaneously sublimate so that the remaining solid reaches 56.4°C, the equilibrium temperature of solid UF_6 at 1 bar.

Globally, the first phase of release of the liquid from the container takes 26.5 seconds, allowing a mass of 9 344 + 86 + 41 = 9 471 kg of gaseous UF_6 to be released, with an average flow of 360 kg/s. The mass of remaining solid UF_6 inside the container is 3 115 – 86 = 3 029 kg.

The next phase is sublimation of the UF_6 remaining in solid form inside the container under the action of the surrounding fire. It is assumed that most of the surface of the container has risen to 650°C (according to the results of <1>). The energy exchanged by radiation between the container and the solid UF_6 serves to sublimate it. It is assumed that the UF_6 is in the form of a cylinder lying on the base of the container. Due to sublimation, the surface of exchange is reduced gradually. It is calculated that the whole of remaining UF_6 sublimates in 6 513 seconds, with an average flow of 0.47 kg/s.

Breach located in gaseous phase

In the configuration where the breach is above the free surface of the liquid, it is assumed that the UF_6 is vacated exclusively in gaseous form.

The first phase is a de-pressurization of the container. The compressed gas vacates via the breach. It is shown that the difference in pressure between inside and outside the container is sufficient so that the critical speed of the gas is reached at the breach. Analysis of the critical speed at the breach shows that the critical mass flow varies little with the internal pressure. The average mass flow between 46 and 12 bar is evaluated at 10.3 kg/s. For a gaseous quantity of 41 kg of UF_6 released, the de-pressurization phase lasts 4 seconds.

In the second phase, the liquid at 150°C that reached the saturation vapor pressure of 12 bar is spontaneously evaporated inside the container. The critical flow of 12.2 kg/s is reached. However, as this will be shown in the next item, the average temperature of the liquid will have a tendency to decrease during vaporization. The pressure inside the container, assumed to be equal to the saturation vapour pressure, will therefore also decrease. It is then verified that if the critical speed at the breach has a tendency to decrease, the mass flow remains essentially constant due to the fact of the increase in the density of the gaseous UF_6. The flow of gaseous UF_6 in this phase of vaporization of the liquid inside the container is therefore approximately equal to 12.2 kg/s. The vaporization phase lasts as long as there is liquid inside the container.

It should be noted that the vaporization reaction being endothermic, the liquid has a tendency to cool in order to provide the energy needed. In fact, it is shown that the power provided by the fire to the liquid via the walls of the container (evaluated at 705 kW) is not sufficient to supply the energy necessary for vaporization (evaluated at 915 kW).

Vaporization is followed by solidification of a part of the liquid at 64°C whilst the complementary part continues to evaporate. The UF_6 being entirely in solid form, a part sublimates so that the remaining solid reaches the temperature of 56.4°C.

The second phase lasts 12 min 07 seconds and allows the release of 8 847 kg of gaseous UF_6 and the formation of 495 kg of additional solid UF_6 inside the container.

The next phase is sublimation of the 3610 kg of solid remaining inside the container. This phase is identical to that detailed in the configuration where the breach is located in liquid phase. The phase of solid sublimation inside the container lasts 6 912 seconds and releases 3 610 kg of UF_6 with an average flow of 0.52 kg/s.

Report on the analysis of the release of gaseous UF_6

It is observed that for each of the two configurations of leakage from the container there are two successive phases: one phase known as « rapid » corresponding to vaporization of the liquid UF_6 and which relates to high level release flows of gaseous UF_6, followed by a phase known as « slow » corresponding to the sublimation of the solid UF_6 and which relates to limited release flows of gaseous UF_6. These two phases are detailed in table 1 and in diagram 2.

Leak configuration from the container		Rapid phase (liquid vaporization)		Slow phase (solid sublimation)
Breach in liquid phase	Duration	27 s		108 min
	Flow (kg/s)	360		0.47
	Released gaseous UF_6 (kg)	9 500		3 000
Breach in gaseous phase	Duration	4 s	12 min	115 min
	Flow (kg/s) Released gaseous UF_6 (kg)	10 41	12 8 850	0.52 3 600

Table 1: 48Y package – Gaseous UF_6 release flows

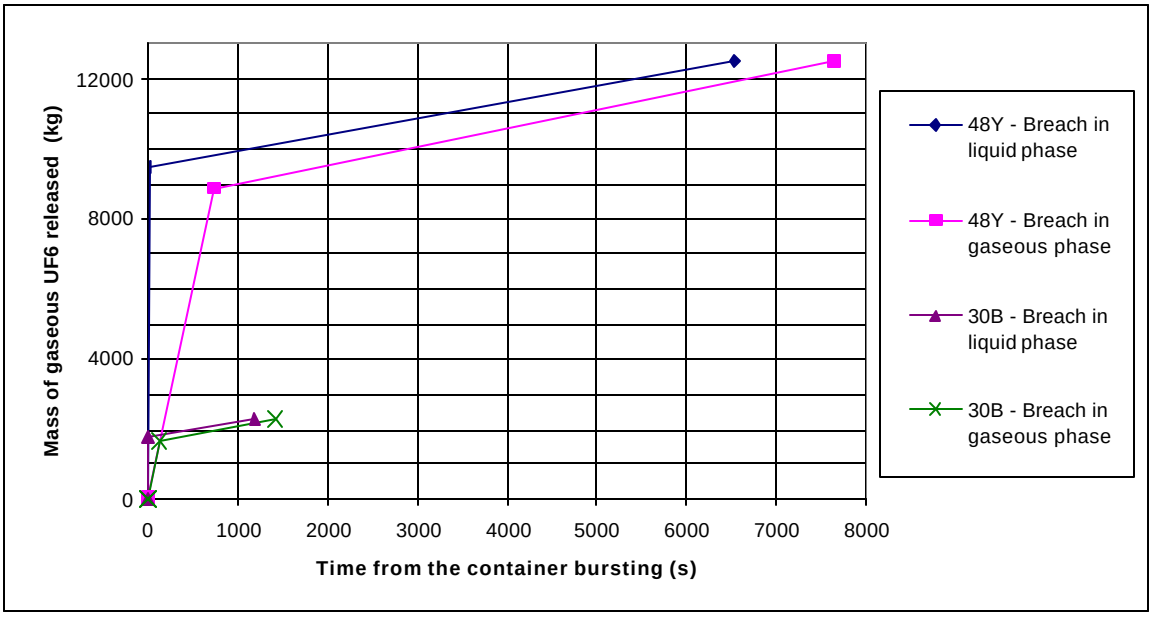

Figure 2: 48Y and 30B packages – Average release of gaseous UF_6 in relation to time

Type 30B package

For comparison, by way of example, the same evaluations have been carried out for a 30B cylinder supposing that at the point of rupture the distribution of the masses for the three phases is identical to those for 48Y. This hypothesis is not truly founded due to the fact that the 30B cylinder is equipped with a thermal protection overpack. Using the same release model, the masses of released gas are indicated in figure 2.

CALCULATION OF THE TOXIC AND RADIOLOGICAL CONSEQUENCES

It is considered that the hydrolysis reaction of the released gaseous UF_6 is total and almost instantaneous. This hypothesis conforms to experience resulting from the observation of accidental release of UF_6 into the open air and from gaseous UF_6 release tests [7]. The mass flow releases of HF and UO_2F_2 are then directly proportional to the flow of UF_6.

Toxic consequences due to the release of UF_6

Four scenarios are considered: Type 48Y or 30B container – Breach in liquid or gaseous phase.

The calculations of the toxic consequences were done based on the IPSN code ICAIR4, considering that the fluohydric acid and the uranium (in the form of UO_2F_2 aerosols) disperse like passive gases.

- Two atmospheric conditions (stability category and wind speed) are considered. They are defined according to the Doury scale ; the relationship with Pasquill's scale is given in brackets: DF3 (f3) and DN6 (c6).
- The uranium deposit speed used is 0.005 m/s ; HF is assumed not to be deposed.
- The calculations are made for ground level releases. The thermal uprise of the plume due to fire could be calculated using the Briggs formula, but the release conditions are too imprecise to obtain exploitable results. Moreover, only thermal uprise more than 10 meters can have significant influence on release calculations.

For each release scenario, three calculations were carried out: an « overestimated » calculation representing the release of all the UF_6 present inside the container at the rapid phase release flow, a calculation that only takes into account the release during the rapid phase (vaporization of the liquid UF_6), a calculation taking into account the rapid phase release followed by the slow phase release (sublimation of the solid UF_6).

Table 2 presents the detail of the results for a type 48Y container where the breach is in liquid phase.
The table contains the results of HF and uranium (in the form UO_2F_2) toxicity analysis.
For each of the components, the atmospheric concentration is given at 500 metres from the release point, in the wind direction. For the uranium, the maximum distances at which the fatal exposure level (level for which the lethal level is 50%) is reached and the level for appearance of renal injury are given. For HF, the maximum distances at which the fatal exposure level is reached and the IDLH are given. The IDLH corresponds to the maximum concentration that can be inhaled without irreversible health effects (cumulated inhalation of HF leading to an integrated amount of 3750 mg in 30 minutes). The respiratory flow used for the evaluation of toxic consequences is equal to 1.2 m^3/hour.
Two meteorological conditions are presented.

Calculation scenario		**HF**			**Uranium (UO_2F_2)**		
		C(HF) max at 500 m (mg/m^3)	**Reaching limit distance (m)**		**C(U) max at 500 m (mg/m^3)**	**Reaching limit distance (m)**	
			Fatal level	**IDLH**		**Fatal level**	**Renal injury level**
DF3	**Overestimated case**	33866	2600	5700	81526	690	4200
	Rapid Phase	28655	2200	4800	68961	590	3700
	Rapid + slow phases	28655	2300	4900	68961	660	4000
DN6	**Overestimated case**	11599	1500	3300	32645	< 500	2500
	Rapid Phase	11391	1400	2900	32060	< 500	2100
	Rapid + slow phases	11391	1400	2900	32060	< 500	2400

Table 2: Type 48Y container – Breach in liquid phase configuration – Consequences of the release of gaseous UF_6

The same calculations have been mde for the three other scenarios. Table 3 presents the reaching limit distance for irreversible health effects related to HF toxicity (IDLH) which is more severe than uranium toxicity (renal injury) for every scenario considered. These are the maximum distances established for the atmospheric condition considered to be the worst (DF3) and considering a realistic configuration: simulation of the rapid phase followed by the slow phase.

Scenario	**Maximum reaching limit distance for irreversible effects**
48Y – Breach in liquid phase	4 900 m
48Y – Breach in gaseous phase	4 200 m
30B – Breach in liquid phase	2 200 m
30B – Breach in gaseous phase	2 000 m

Table 3: Toxicity of HF and Uranium – Atmospheric condition DF3

The study of the configurations considered raises the following points:

- the scenario concerning the type 48Y container naturally has greater toxic consequences than the scenario involving the type 30B container,
- the configuration « Breach in liquid phase » leads to more damaging results than the configuration Breach in gaseous phase » ; however the extent of the consequences for the two scenarios are the same,
- the maximum toxic effect, for the fluohydric acid as well as for the uranium, is reached in its almost totality during the rapid phase that relates to the vaporization of the liquid UF_6. The atmospheric concentrations only increase very slightly during the sublimation phase, which is explained by the low level flows brought into action during this phase,

- the reaching limit distances for toxic level leading to HF irreversible health effects are greater than the reaching distances for the toxic level related to uranium.

Radiological consequences due to the release of UF_6

Calculations of the radiological consequences due to the release of UF_6 were carried out using the IPSN SIROCCO code that integrates the Doury atmospheric dispersion models with the DF3 and DN6 meteorological conditions for a release at ground level.

The calculations only concern the type 30B container containing UF_6 enriched with 5% Uranium U235, the rate from which the radiological consequences of uranium release may become more damaging than its toxic consequences.

The calculation made relates to 1 555 kg of uranium, corresponding to 2 300 kg of UF_6, released in 10 minutes (the minimum duration imposed by the code).

The specific contents used for each isotope of uranium are as follows (in g/gU): U232: 1.10^{-10} **;** U234: 5.10^{-4} ; U235: 5.10^{-2}; U236: $2{,}5.10^{-4}$; U238: $9{,}5.10^{-1}$.

The code allows calculation of the total effective dose for an adult following 24 hour exposure time as a function of the distance to the release point in the wind direction. In table 4, the calculated reaching limit distances for levels of 50 mSv and 10 mSv are deduced. These levels are respectively recommended in France by the "Direction Générale de la Santé" (public health administration) for evacuation and shielding of the population.

Level ↓	**Meteorological condition ⊗**	**DF3**	**DN6**
50 mSv exposure level		620 m	< 500 m
10 mSv exposure level		1580 m	830 m

Table 4: distance to the release point allowing the exposure level given to be reached depending on meteorological conditions

If we compare the reaching distances for the 10 mSv level with the reaching distances for the renal injury level in the most damaging configuration (1 800 m and 960 m respectively in DF3 and DN6), it can be noted that the toxic consequences of the uranium release enriched at 5% are more severe than its radiological consequences: the reaching limit distance for irreversible health effects is shorter in the case of toxic effects.

In general, it is noted that the toxic consequences of the HF are greater than toxic and radiological consequences of the uranium.

CONCLUSION

The toxic effects of the HF from the hydrolysis reaction of the UF_6 are preponderant on the toxic or radiological effects of the UO_2F_2 in the case where the enrichment of UF_6 is less than 5%.

The analysis carried out shows that the UF_6 releases from a type 48Y or a type 30B full container exposed to a long duration fire leading to the bursting of the container may have irreversible effects for populations up to 4 900 m from the release point within the meteorological conditions used. The reaching distance for 50% lethal consequences is, according to the analysis, 2 300 m.

For package design approved according to a H(M) certificate (this would be the case for type 48Y containers not equipped with fire protection for which the risks of rupture due to a severe fire are more probable), the feasibility of setting up in approximately half an hour a security perimeter located at 5 kilometres from the accident should be considered.

In addition, it remains to be confirmed that the possible leakages of UF_6 from the valve and plug connections of a 48Y container, that occur a long time before the rupture, have a sufficiently limited flow so that a safety perimeter placed at least 500 metres from the container is adequate.

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