
Radiological consequence analysis in case of fire impact

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Abstract:

Airborne release and dispersion of radioactive material from objects involved in a fire scenario includes a combination of coupled processes that may as well amplify as counteract in terms of consequences in the environment. Some of these relevant processes and parameters are type of release (spontaneous or continuous), time of release, thermal lift, spatial and temporal variation of fire environment and spatial distribution of objects. An exemplary sensitivity study of the behaviour of a set of 48Y cylinders filled with UF₆ exposed to a large hydrocarbon fire is presented. The analysis reveals that a detailed consideration of process interaction and parameter variation is needed to identify most unfavourable event sequences with respect to radiological and chemotoxic consequences.

1 INTRODUCTION

Severe fire scenarios with a significant release of radioactive material due to direct impact of internal fires are rather unlikely in nuclear facilities since combustible material is limited to a minimum. However, the impact of externally induced fires, particularly on packages used for transport and storage of radioactive material, may cause a release of radioactivity to the environment. Therefore, this paper focuses on severe fire scenarios due to accidents or malevolent actions where transport or storage packages with radioactive material are involved. Although not probable, these kinds of scenarios may imply the potential of a significant release of radioactive material to the environment.

In the past several studies have been performed on this topic at GRS for various types of packages involved in the fuel cycle as well as for interim and final storage of waste and spent fuel casks. Within the last years some of these studies also included three-dimensional CFD (Computational Fluid Dynamics) fire modelling in order to derive detailed information about potential temperature and heat flux pattern to engulfed objects. With this type of model also complex configurations of objects, fire area and ventilation can be analysed. Fig 1.1 gives an example of a benchmark simulation with the CFD fire model FDS4 (Fire Dynamics Simulator [1]) for a waste package partially engulfed by a kerosene pool fire.

Apart from the application of suitable tools to analyse the package performance in fires it is essential to identify the dominant scenario with respect to the severity of radiological consequences among the wide range of potential scenarios. Various phenomena have to be taken into account depending on the scenario and on the packages involved. Within the following sections the importance of some phenomena which influence the release and the dispersion of radioactive material in fire scenarios will be discussed in general. In order to

discuss some of these aspects in more detail an example analysis of a fire scenario with a release from UF₆ cylinders is given.

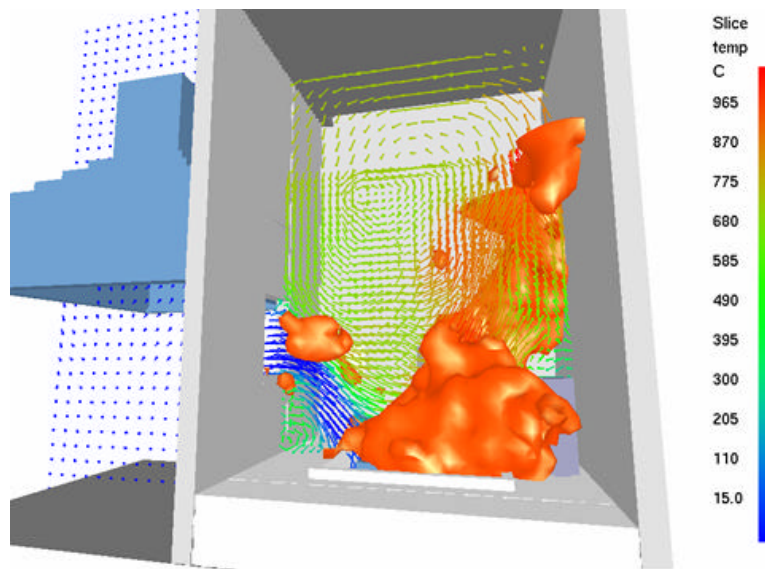


Fig. 1.1: Example of the flame and flow pattern near to a waste drum (right) in a ventilated compartment simulated with FDS4 (color scale gives gas temperatures)

2 RELEASE AND DISPERSION PHENOMENA

Depending on the actual accident or sabotage scenario up to four fire environments with different release and dispersion characteristics can be defined:

- initial fireball
- spreading and growing fire
- fully developed fire
- post fire environment.

A fireball may occur in collision scenarios with liquid or gaseous fuel, e.g. due to intentional impact of an aircraft or due to an accidental collision with a tank car. Immediate mechanically driven release of airborne radioactive material will experience a strong updraft and rapid dilution due to the build-up and rise of the fireball and the remaining gases up to several hundreds of meters depending on the fuel mass and on atmospheric stability. Therefore, even substantial release during this phase may be of less severity compared to the following phases if a fireball occurs.

When the fireball has disappeared or in cases without an initial fireball any further early phase airborne release from packages while the fire is spreading and growing will experience a much lower plume rise implying a higher potential threat to the downwind area relative to the fireball environment due to near ground dispersion Fig. 2.1 shows exemplary FDS4 results of the thermal lift of near ground particle release in a fireball environment (left and center) and in a subsequent pool fire. Results generally showed good agreement with plume rise models as [2] or [3].

With respect to thermally driven airborne release of radioactive material (e.g. evaporation, pyrolysis, aerosols from burning etc.) the fire spreading and growing phase is of lower importance compared to the subsequent phase of a fully developed fire due to the thermal

inertia of packages and of the radioactive material itself. On the other hand, the thermally driven plume rise increases with increasing heat release rate of the fire, commonly reaching heights of several hundreds of meters for fires with 100 MW order magnitude the potential to trigger significant releases by thermal impact [3]. Hence, it may not be obvious at the start of an analysis which of both fire phases could be more severe with respect to radiological consequences.

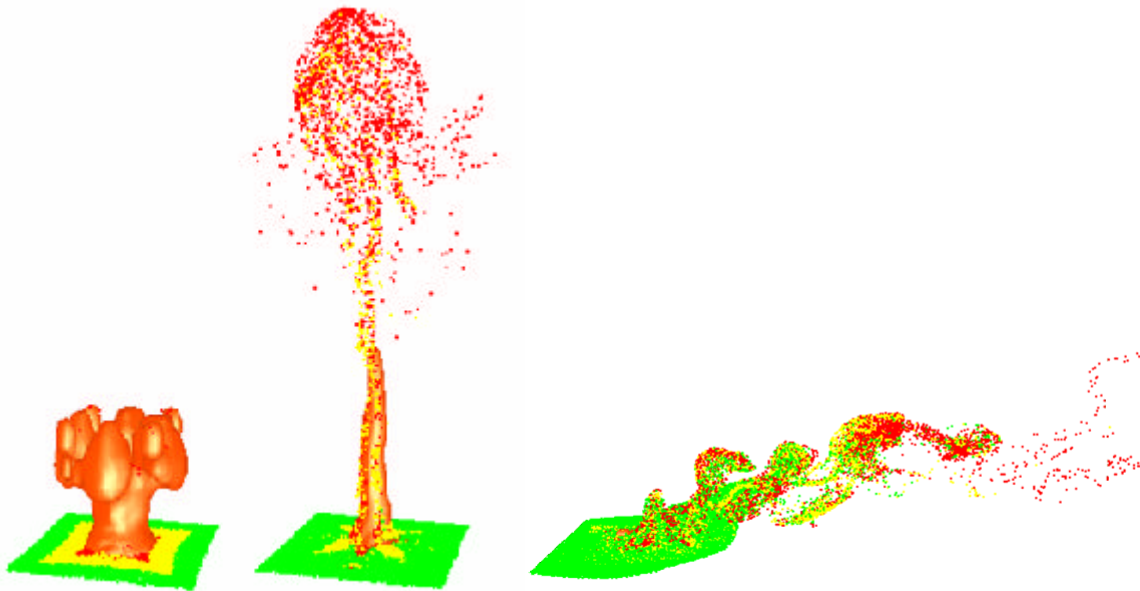


Fig. 2.1: Example of flame pattern and particle dispersion of near surface release during a fireball phase (left and center: developing fireball and remaining particle cloud) and a subsequently spreading and growing pool fire (right) simulated with FDS4

Furthermore, the thermal inertia of packages may lead to a delayed release of material during the post fire phase. This delayed post fire release may either be due to a temperature wave still propagating into the radioactive material with diminishing amplitude, continuing depressurisation via a comparably small leak or even a rupture due to ongoing internal pressure build-up after the end of the fire. There will be only a minor thermal lift during this phase which is driven by hot objects and by residual fire nests or smouldering.

Hence, the careful analysis of the sequence of events may be crucial for radiological dose calculations. The temporal development of events itself may be subject to large variability. Thus, even if the amount of combustible material involved in the fire is known it is still difficult to decide which scenario will dominate the radiological consequences without analyzing some branches of the scenario event tree in detail. This also holds for further parameters of the fire scenario as fire extent, heat release rate and spatial distribution of objects which will be discussed in the following section.

3 THERMAL PERFORMANCE OF OBJECTS

Given the amount of combustible material involved in a fire and its energy content there may still be a range of potential scenarios of heat release (fire temperature) and respective fire duration. As an example, Fig. 3.1 gives the temperature development in a steel package wall for two engulfing fires which are nearly equivalent with respect to their integral (radiative) net heat flux to the package:

- the standard test fire for a Type B Package: 30 min, 800°C

- a large hydrocarbon fire of 1150°C and 10 min duration.

The temperature 1150°C is primarily chosen to obtain a simple factor of 3 between both radiative fluxes. However, it represents a typical value of temperature maxima measured inside large outdoor hydrocarbon fires, e.g [4]. Maximum temperatures within the outer few centimetres of the package wall engulfed by these fires are much higher for the short hydrocarbon fire which may distort the package body in a way that the seal tightness is reduced. Deeper inside the package wall the maximum temperature is similar for both scenarios. In both cases maximum temperatures are reached several minutes after the end of the fire with a slightly longer delay of the maximum for the short fire. Hence, the choice between both fire scenarios basically affects the performance of the outer package region, whereas the thermal impact to the interior is mainly driven by the integral heat flux to the package.

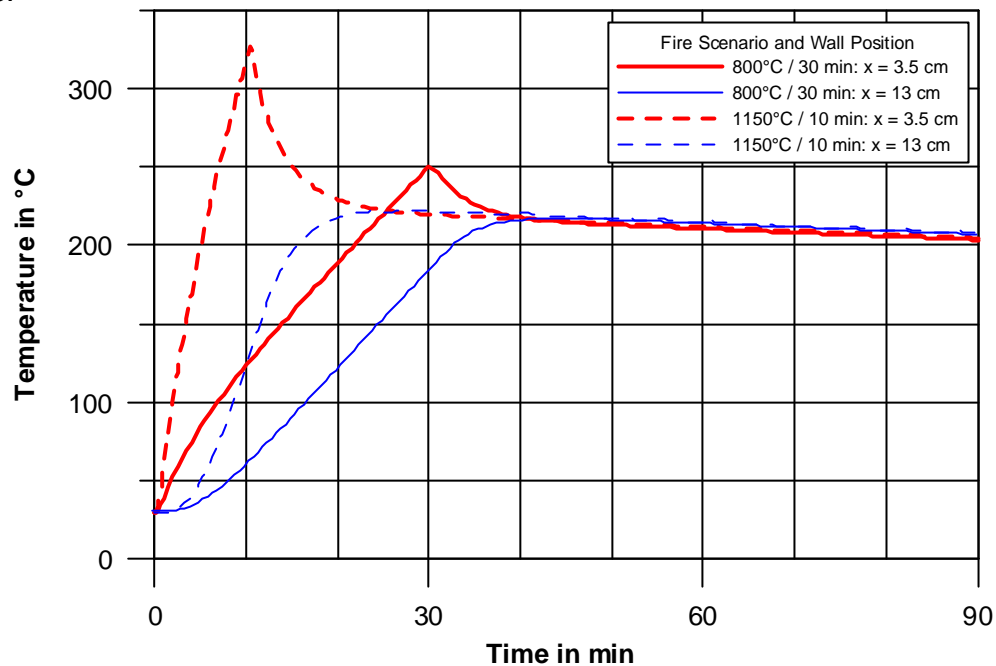


Fig. 3.1: Evolution of simulated package wall temperatures (cast iron) at different depth and for different fire scenarios

The integral heat flux is strongly influenced by the type of exposure of the package to the fire. Obviously, a fully engulfing fire is much more severe for a package than a partly engulfing fire or a fire beneath a package. As an example, Fig. 3.2 shows the dependency of the maximum radiative heat flux to a unit area outside the fire in relation to the heat flux emitted by the unit area of the fire surface (approximated by a cylinder of radius r and height h). This so called viewfactor shows a rapid decrease with increasing distance to the fire. This factor is even higher when comparing it with a package fully engulfed by a fire with approximately four times the package area affected by the fire.

Hence, there is a wide range of thermal impact scenarios for individual objects in the same fire scenario due to

- distribution of objects inside and outside the fire
- temperature pattern of the fire
- temporal development of the fire (area and intensity).

The interplay of thermally induced damage to packages and release on the one hand and the thermal lift of released material the other hand can lead to a broad and complex spectrum of consequences, especially if several packages are involved. There is not necessarily a monotonous relationship between fire parameters and consequences. Instead, there may be an unfavourable combination of fire parameters that may lead to the most severe

consequences which can only be established by analysis of the effect caused by the processes mentioned above.

In the following section, an example of such a scenario is given by calculation of potential radiological and chemotoxic consequences due to a release from UF_6 packages in a hydrocarbon pool fire. The example demonstrates the effect of a realistic parameter variation within one scenario compared to a fixed parameter scenario. Due to the special physical and chemical properties of UF_6 , this example is more complex than for most other radioactive materials.

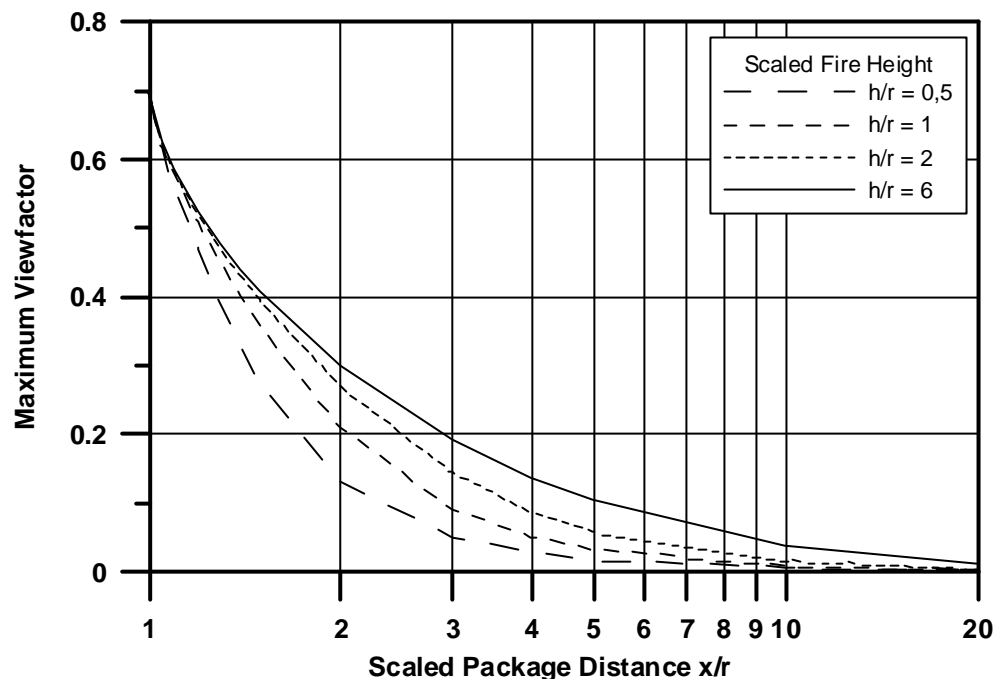


Fig. 3.2: Maximum viewfactors between a fire in cylindrical approximation (height h , radius r) and a plane outside the fire in distance r from the fire center

4 EXAMPLE: BEHAVIOUR OF UF_6 -CONTAINERS IN A POOL FIRE

As an example, the behaviour of ten 48Y cylinders, each containing 12,5 t feed (natural uranium hexafluoride, UF_6), exposed to a large hydrocarbon fire is considered by a sensitivity study. This highly hypothetical scenario is chosen to demonstrate some effects which can be relevant to a variety of situations in which other packages and radioactive materials may be involved.

The effect of variations in fire duration and intensity on the risk of rupture and subsequent elevated and near-surface release is investigated by a sensitivity study. For this purpose, we rely on analyses of the behaviour of a single 48Y container in an engulfing 800° fire by state-of-the-art methods which are well documented in literature (e.g. [5], [6], [7]) and summarized in the following subsection. In order to extend these findings to a release scenario including several containers, we introduce additional assumptions on container behaviour which are described and motivated in subsection 4.2 The results of the sensitivity study of release are described in subsection 4.3 while the radiological and chemotoxic consequences of a representative release scenario are analysed in subsection 4.4.

The details of the rupture and release scenario are closely linked to the properties of UF_6 . Being in the solid state at normal environmental temperature and atmospheric pressure, it sublimates at $56,5^\circ\text{C}$ when heated. A subsequent increase in vapour pressure and temperature by continuous heating causes the material to start liquefying when its triple point (at $0,15\text{ MPa}$ and 64°C) is reached. Further temperature and pressure increase by ongoing heating than may eventually cause the rupture of the container and subsequent flashing of the over-pressured liquefied part of its content. After release UF_6 will rapidly react with the air moisture and the hydrocarbons of the fuel (cf. subsection 4.4 for details). The produced uranium compounds and hydrogen will determine the consequences in some downwind distance.

4.1 Rupture and release scenario for a single 48Y container in an 800°C fire

The behaviour of a single 48Y container is well investigated by a combination of by the TENERIFE heating experiments with numerical simulations by the DIBONA code (cf. [5]) and the PEECHEUR rupture tests (e.g. [6]). These investigations establish a lower bound for rupture of about 26-28 minutes if exposed to an engulfing 800°C fire by heating of the UF_6 and corresponding phase transitions and pressure build-up.

The release of UF_6 from a ruptured 48Y container exposed to a long duration fire has been analysed by [7]. The authors present two different scenarios according to whether the breach is located in the gaseous phase in the upper part of the container or in the liquid phase in the lower part. As the latter is by far more improbable, we rely on the results given in [7] for a location of the breach in the gas phase. According to [7], two release phases can be distinguished:

- In the first phase (denoted “rapid phase” in [7]) the container pressure is reduced to environmental levels by the evacuation of the gaseous UF_6 content and the flashing (i.e partly vaporization of the over-pressured liquid UF_6 content. According to [7], the gaseous content (41 kg UF_6) vacates through the breach at a release rate of 10.3 kg/s within the first 4 seconds. Subsequently, a total of 8850 kg of the initially liquid content evaporates in the next 12 minutes at a release rate of approximately 12.2 kg/s .
- In the second phase (denoted “slow phase” in [7]) the remaining 3600 kg of solid UF_6 sublimate at environmental pressure. Sublimation energy is by and large provided from the fire by irradiative heat transfer that is gradually reduced with the diminishing surface of the solid UF_6 in the container. According to [7], this process takes 115 minutes at an average flow $0,52\text{ kg/s}$. No distinction between fire condition and post fire condition are made in [7],

For our investigation, we adopt the results of [7] for the first phase which is dominated by the flash process but make a different approach for sublimation process the second phase. If the fire lasts longer than it takes the container to rupture plus the depressurization time of the container we assume that the total heat transfer to the container is converted to sublimation energy, leading to release rates of about 10 kg/s until either the fire has ended or the container is empty. As we will show below, this conservative estimate of release by sublimation during fire does not substantially influence our results in terms of near-ground concentrations and doses because the effect of thermal lift on dispersion of released quantities is taken into account. After the end of the fire, the quantities of solid UF_6 released by sublimation are small because ventilation of the container is poor and the UF_6 surface tends to be inactivated by conversion to UO_2F_2 via hydrolysis.

4.1.1 Relevance of release height and fire duration

In [7], the calculation of consequences is based on the assumption that the release is near-ground regardless the thermal lift by the fire. This assumption tends to provoke a conservative overestimate of effects as near ground concentrations and doses of UF_6 reaction products are substantially reduced if thermal lifts of several 100 meters are taken into account. As mentioned in section 2, a fire which is vigorous enough to cause a 48Y container rupture also leads to thermal lifts which are at least of this order of magnitude. The counteracting effect of larger molecular weight of UF_6 and chemically converted uranium compounds on buoyancy is only of minor importance and partly overrun by additional heat production due to the reaction of UF_6 with hydrocarbons. We thus assume a release height of 400 m while the fire is burning in its fully developed stage.

It is possible that the fire substantially weakens and finally dies before the release due to depressurisation and flashing of the liquefied content has ended. The release height will then be influenced by the complex interaction of the mechanical momentum of the flow, the heavy molecular weight of the UF_6 gas and reaction products, heating from hot surfaces and the chemical conversion to HF and UO_2F_2 by hydrolysis and the turbulent mixing with ambient air. As we are interested in the effects at some distance from the source, we mimic the net effect of these complex interactions by a volume source with a vertical extension of 10 m and a center at about 10 m height. The longer the fire lasts after rupture, the smaller is the quantity of near-ground release. The relation between rupture time and fire duration thus exerts a substantial influence on the consequences.

4.2 Rupture sequence for several 48Y containers in a fire with varying temperature

If several UF_6 containers burst by thermal impact of a large fire of long duration, the superposition of the release from each container will determine the total release. The time it takes for each container to rupture will vary with the amount of radiation energy absorbed, differences in heat transfer and phase transitions of the contents and the variations in the properties of the container steel. The simplified assumption of an instantaneous rupture of all containers at once would provoke an overestimation of the net release flow and an inappropriate sensitivity of the consequence analysis on the duration of the fire. It is thus necessary to establish an estimate of the time interval between the rupture of the first and the last container (denoted rupture time interval hereafter). The lower bound of this time interval for a homogeneous, engulfing 800°C fire can be adopted from the results of the TENERIFE tests and corresponding DIBONA calculations [5] to be about 26 minutes in an 800°C fire. The upper bound is subject to large uncertainty. As pointed out by Baze et al., 2001 [8], rupture time is sensitive to the mechanisms of heat transfer within the UF_6 content and can exceed half an hour. We estimate the upper bound to be 35 minutes for an engulfing 800°C fire.

As mentioned in section 3, fire temperatures can be substantially higher than 800°C in a large hydrocarbon fire and the spatial distribution of temperature is not homogeneous but undergoes considerable variations. The spatially inhomogeneous temperature distribution within the fire in turn alters the radiant heat transfer to the containers. Using the Stefan-Boltzmann law, we estimate the dependence of rupture time on fire temperature by assuming that rupture takes place when the container has received the same amount of radiant heat as within the rupture time established for the 800°C fire. The appropriateness of this simple scaling method is supported by the results presented in Fig 3.1 and by estimates of 48Y container rupture times based on comparison between the energy needed to reach critical pressure in the container and radiant heat transfer to the container (e.g. [9]), which are in good agreement with the results from the sophisticated methods cited above (e.g. [5], [6]). Though not being applied here, the net effect of reduced heat transfer to containers which

are not fully engulfed by the fire (cf. section 3) could be estimated by a similar scaling approach.

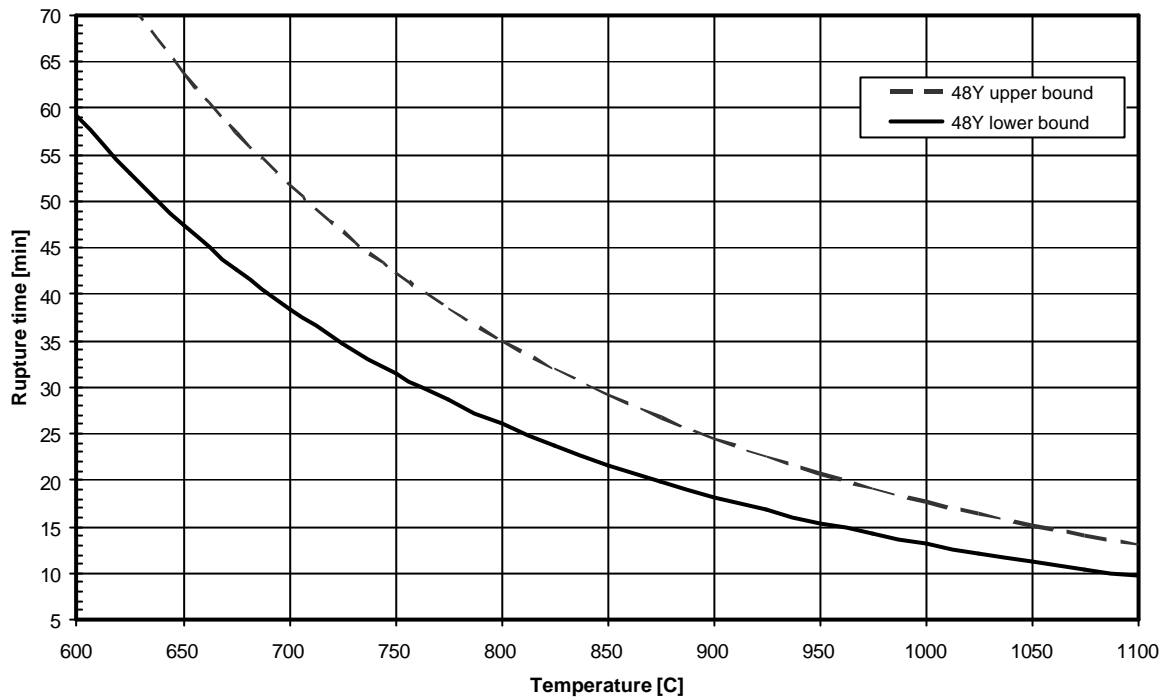


Fig. 4.1: Dependence of rupture time interval on flame temperature for a 48Y container

The estimate of the dependence of the rupture time interval on flame temperature (Fig. 4.1) shows considerable sensitivity of rupture times to the intensity of the fire. Hence, the difference between rupture times of several containers tends to increase if the containers are exposed to different flame temperatures. Taking into account temperature variations thus leads to a broadening of the rupture time interval.

As there is no information available on the distribution of rupture times, we assume the rupture times to be uniformly distributed within the respective time interval. By this, an individual rupture time is attributed to each container. We assume a container to rupture if the rupture time is shorter than the fire duration plus a margin of 100 seconds which accounts for the possibility of ongoing pressure build-up after external heating has stopped. This possibility is known from the TENERIFE tests [5].

4.3 Dependence of release from bursting containers on fire duration and intensity

Based on the methodology described in subsections 4.1 and 4.2, release sequences for fire durations between 15 and 45 minutes are computed. In order to reveal the difference between homogeneous and inhomogeneous heating by the fire, two fire scenarios are compared. For scenario 1, a spatially constant flame temperature of 800°C is assumed in the whole area. Scenario 2 is based on a linear temperature variation between 800°C and 1000°C in the affected area.

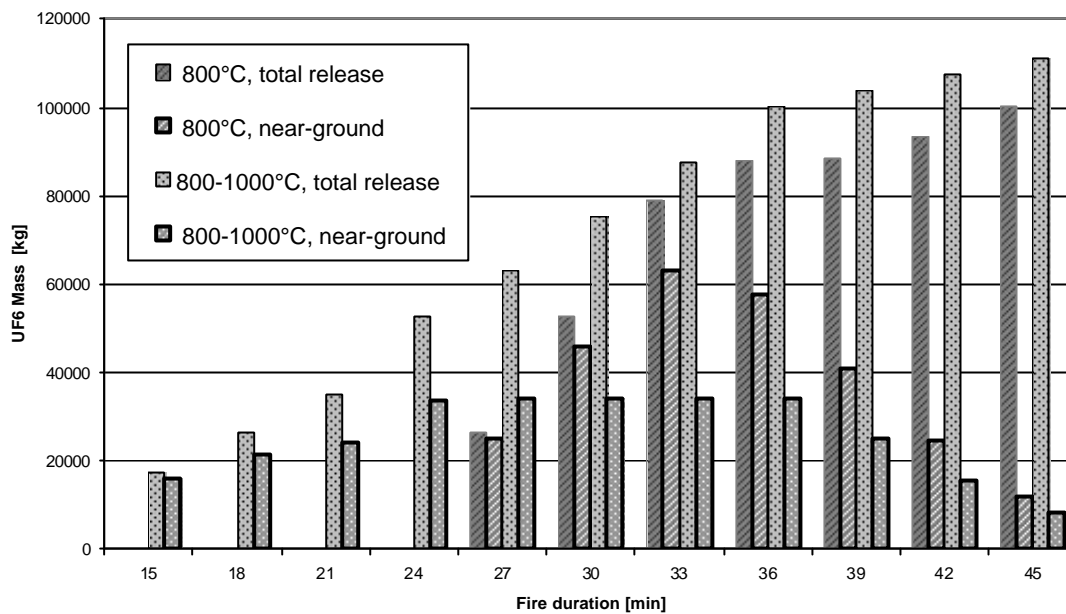


Fig. 4.2: Total and near-ground quantities of UF_6 released for different fire durations and two scenarios of flame temperature.

Resulting releases are depicted in Fig. 4.2. It is evident that the total quantities released increase with growing fire duration for both scenarios. The quantities of UF_6 released near-ground show a different behaviour, peaking for fire durations of about half an hour. This is due to the fact that with increasing fire duration the number of containers that rupture grows but the portion of UF_6 that is thermally lifted also increases. Moreover, the maximum near-ground release is smaller for scenario 2 than for scenario 1 and the sensitivity of near ground-release to fire duration is less pronounced than in scenario 1. It is evident that inclusion of temperature variations in the analysis leads to a reduction of near-ground releases in spite of the effect of higher flame temperatures. As near-ground released quantities dominate the radiological and chemotoxic consequences, the effect of uncertainties in the knowledge of fire duration is also reduced by the inclusion of temperature variations in the fire.

4.4 Analysis of radiological and chemotoxic consequences

Radiological and chemotoxic consequences are investigated for a fire duration of thirty minutes for fire scenario 2 (spatial temperature variation between $800^\circ C$ and $1000^\circ C$). After release UF_6 reacts rapidly with either the moisture of the air to HF and UO_2F_2 or with the hydrocarbons of the fuel. The complex reaction chemistry within the fire can lead to the formation of a variety of products (e.g. [9]), of which the uranium compounds and HF dominate the consequences. For simplicity, we assume that, during the fire, UF_6 is predominantly converted to UF_4 and HF while it is hydrolysed to HF and UO_2F_2 when released after the fire. Atmospheric dispersion of the reaction products is computed with the particle model LASAT, which is extensively tested and widely employed at GRS for a broad range of applications. Turbulence parameters are derived by the procedure established in [10] based on Monin-Oboukhov boundary-layer theory. By this method, a validated parameterisation of turbulence which yields physically consistent parameters for near-ground as well as for elevated release is applied. Near-ground concentrations of Uranium and effective doses are calculated for a stable (Pasquill diffusion class E), neutral (class D) and

unstable (class B) weather situation with weak winds (1 m/s in 10 m height) and medium surface roughness. A deposition velocity of 0,0015 m/s is assumed for uranium compounds.

The time development of near-ground uranium concentration is illustrated by Fig. 4.3 for the most unfavourable stable situation at three downwind distances of 1000 m, 2000 m and 5000 m. A comparison with the AEGL (Acute Exposure Guideline Level)-Scale for UF_6 (used as estimate for the combined effects of uranium compounds and HF) demonstrates the severity of chemotoxic effects already known from earlier studies (e.g. [7]). The AEGL-3 level for ten minutes exposure, marking the threshold for life-threatening health effects, is exceeded threefold even at a distance of 5 km. Only at distances of about 8 km this concentration level is no longer met.

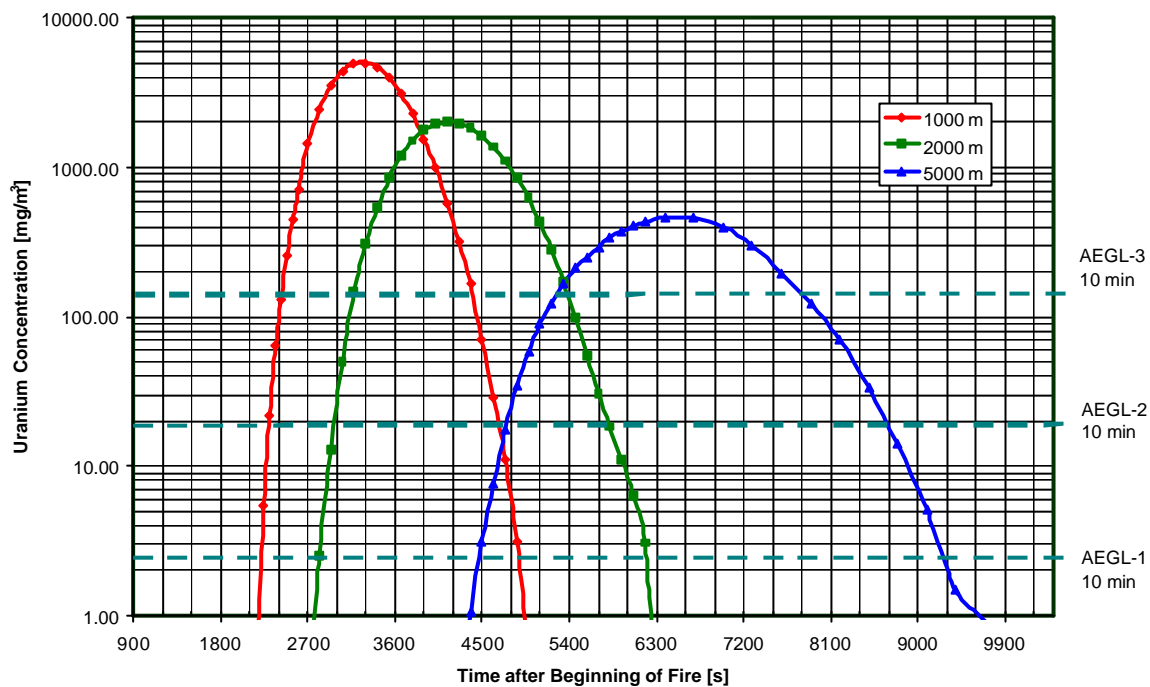


Fig. 4.3: Time development of uranium at three distances from release concentrations for a stable weather situation. The curves represent 10 minute running averages, thus being directly comparable to the corresponding AEGL values referring to the uranium share in UF_6 .

Chemotoxic effects dominate the radiological impact. This is evident by considering the effective inhalation doses for adults presented in Fig. 4.4. The value of 100 mSv, for example, which would justify incisive countermeasures such as evacuation according to German regulation, is not met. Moreover, the higher doses in shorter distances are somewhat hypothetical as the toxic damage to the breath apparatus by the HF alone would make it impossible for any individual to inhale the corresponding quantities of uranium compounds.

The material released at higher levels during the fire contributes notably to dose only for the unstable situation at distances larger than about 1500 m. This contribution from higher elevations is visible by the reduced steepness of dose decrease with distance compared to the other weather situations. Further analysis reveals that, for the unstable weather situation, contributions from elevated release add no more than 0.3 mSv to the entire effective dose (not shown). The dominance of near-ground releases is thus confirmed for all weather situations.

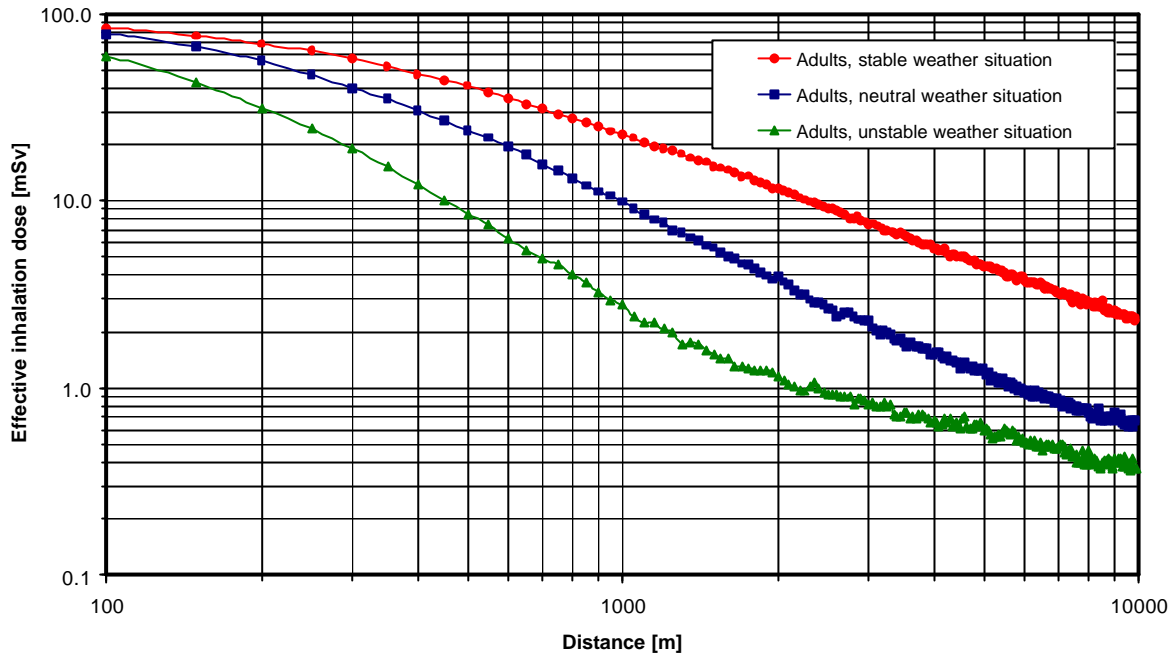


Fig. 4.4: Effective inhalation dose for adults for three different weather situations

5 CONCLUSIONS AND OUTLOOK

The results of our sensitivity study demonstrate that the net effect of a fire on damage the release of radioactive material from leaking packages is determined by a combination of coupled processes that may as well amplify as counteract. Among these processes, the most relevant are

- possibility and expected time of damage to the packages
- thermally induced release
- thermal lift by the fire
- possibility and duration of ongoing release after the fire has ended.

The example presented shows that it is only the consideration of process interplay which leads to a reasonable identification of most unfavourable event sequences. For example, the assumption that the longer and hotter the fire, the more severe the consequences would be, has been demonstrated to be incorrect. Instead, unfavourable combinations of fire duration and intensity could be identified.

In particular, the timeline of package damage and release has to be carefully established to identify unfavourable scenarios as well as to avoid unrealistic overestimation of the consequences. If a large area with several packages is involved, it is also advisable to take spatial inhomogeneities in fire conditions into account to reduce overestimation of consequences as well as uncertainties associated with the limited knowledge of the fire parameters for a real situation. Supporting information on spatial and temporal variation of fire scenarios can be obtained by suitable analysis of fire experiments and CFD fire model results.

These findings also apply to analysis of release scenarios with e.g. leaking cast iron packages in a fire. The release will be influenced by the pressure build up within the package by heating, which is in turn delayed with respect to the forcing by the fire by the time it takes

for the heat wave to move into the cask. Hence, the possibility of significant post-fire near-ground release cannot be excluded and a detailed analysis is necessary to determine the consequences and identify the most unfavourable conditions. Further analyses within this context will be carried out at GRS.

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