Experiments to quantify airborne release from packages with dispersible radioactive materials under accident conditions

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Abstract:
For transport or handling accidents involving packages with radioactive materials and the assessment of potential radiological consequences, for the review of current requirements of the IAEA Transport Regulations, and for their possible further development reliable release data following mechanical impact are required. Within this context a research project was carried out which extends the basis for a well-founded examination of the contemporary system of requirements of “Low Specific Activity” (LSA)-type materials and allows for its further development where appropriate. This project comprises a prior system-analytical examination and an experimental programme aiming at improving the general physical understanding of the release process as well as the quantity and the characteristics of airborne released material for non-fixed dispersible LSA-II material upon mechanical impact. Impaction experiments applying small, medium and real sized specimens of different dispersible materials revealed that the release behaviour of dispersible powders strongly depends upon material properties, e.g. particle size distribution and cohesion forces. The highest experimentally determined release fraction of respirable mass (AED < 10 \( \mu \text{m} \)) amounted to about 2 % and was obtained for 2 kg of uncontained easily dispersible pulverised fly ash (PFA). For larger uncontained PFA specimen the release fraction decreases. However, packaging containing powdery material substantially reduces the airborne release fraction. The measured airborne release fractions for a 200 l drum with Type A certificate containing PFA were about a factor of 50 to 100 lower than for uncontained material. For a drop height of 9 m the airborne release fraction amounted to about 4 \( \times 10^{-5} \). This value should be applicable for most of transport and handling accidents with mechanical impact. For a metal container of Type IP-2 or better which contains powder masses of 100 kg or more this release fraction may be adopted after a 9 m drop.

Based on these experimentally determined release fractions for packages containing LSA-II material in powder form it can be concluded that the resulting inhalation dose of an individual in the vicinity of the accident location lies by more than one order of magnitude below the effective dose limit of 50 mSv. Thus, the limits specified in the Transport Regulations of the mass-related activity concentrations for LSA-II of \( 10^{-5} \cdot A_{239}/g \) were confirmed as being conservative also for powdery, easily dispersible materials.

1 INTRODUCTION

The IAEA Regulations for the Safe Transport of Radioactive Materials \cite{IAEA} regulate the transport of packages containing “Low Specific Activity” (LSA)-type materials. Requirements concerning the material characteristics of the allowed radioactive contents and the quality of the packaging are mainly derived from considerations of potential radiological consequences...
to individuals following severe transport or handling accidents. Of major concern are airborne releases of radioactive material as consequence of an accidental impact which can result in a radiation exposure of persons in the vicinity of an accident site via inhalation of radioactive particulate.

The Transport Regulations distinguish three types of solid radioactive LSA-materials:
- LSA-I: essentially uranium and thorium ores and concentrates of such ores as well as natural or depleted uranium or natural thorium
- LSA-II: material in which the activity is distributed throughout and the estimated average activity concentration does not exceed \(10^{-4} \cdot A_2/g\) (\(A_2\) being the radionuclide-specific activity limit for Type A packages for material which does not qualify as special form). A large fraction of LSA-II packages contain various kinds of radioactive wastes in solid form and may include powdery materials.
- LSA-III: Solids (e.g. consolidated wastes), excluding powders, in which the radioactive material is distributed throughout a solid or a collection of solid objects, or is essentially uniformly distributed in a solid compact binding agent (such as concrete, bitumen, ceramic) and the estimated average activity concentration of the solid does not exceed \(2 \times 10^{-3} A_2/g\).

For transport or handling accidents involving packages with radioactive materials and the assessment of potential radiological consequences, for the review of current requirements of the Transport Regulations, and for their possible further development reliable release data following mechanical impact are required. This is definitely one of the demanding issues in the field of transport safety of radioactive materials.

For radioactive substances which are either embedded in brittle matrix material (e.g. in cement, glass, or ceramic) or in a dispersible form the amount of accidentally generated airborne particulate matter and its particle size distribution as a function of aerodynamic diameter is of special importance. In most accident situations the radiological consequences are dominated by the aerosol release. Particles up to about 10 \(\mu m\) aerodynamic equivalent diameter (AED) in size are respirable and can reach deeper regions of the lung, where clearance times may be long. Particles between 10 \(\mu m\) and 100 \(\mu m\) AED are of less concern for the inhalation pathway, but they can contribute to other exposure pathways after deposition. Particles greater than 100 \(\mu m\) AED deposit very quickly and are therefore of concern only in the immediate vicinity of the location of release.

By the example of radioactive wastes immobilised in brittle materials, e.g. cement, concrete, glass, ceramics or other brittle materials such as fresh fuel, recently the release behaviour of non-fixed LSA-III materials upon mechanical loading was studied with respect to the fragmentation and the release behaviour of brittle material under mechanical loads. This investigation was based on a prior system-analytical examination and comprised an experimental programme aiming at improving the general physical understanding of the release process as well as the quantity and the quality of release data. By combining laboratory experiments using small scale test specimens with a few key scaling experiments with large scale test objects significant progress was achieved to meet this objective. The laboratory equipment enabled the in-situ determination of the amount and aerodynamic size distribution of the airborne particles generated upon impact of the test specimen on a hard target. Impact energies covered the range experienced in transport accidents including aircraft accidents. The well defined experimental boundary conditions and the good reproducibility of the experimental procedure allowed for systematic studies to measure with good precision the amount and aerodynamic size distribution of the airborne release and to quantify its dependence on relevant parameters such as energy input, material properties, specimen geometry. The experimental programme was performed within the scope of various national and international (e.g. EU-funded [2], [3]) projects.
The small scale experiments with brittle materials revealed a pronounced universality of the airborne release in view of the material properties and the aerodynamic size distribution. These results form a valuable data base to limit the number of key large scale experiments aiming at extrapolation to full size realistic packages. They also justify the use of a surrogate material in these tests so that the release fractions determined for this specific material are representative for a wide class of brittle radioactive materials.

Because of the outstanding importance of reliable quantitative data for accidentally generated airborne particulate matter from packages containing LSA material the investigations were continued with respect to non-fixed dispersible LSA-II material upon mechanical impact. A corresponding research project - funded by the Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (BMU) and the Federal Office of Radiation Protection (BfS) - was carried out. This project extends the basis for a well-founded examination of the contemporary system of requirements of LSA material and allows for its further development where appropriate.

The central part of the project was the performance of an experimental programme with the aim to determine the influence of energy input, package size, packaging and material properties (like e.g. particle size distribution, cohesion forces) on the release of airborne dusts from packages containing easily dispersible material upon their impact on unyielding surfaces. The experimental programme comprised large scale drop experiments and small scale impaction experiments which are described in Section 3.

In the following Sections the selection of appropriate material systems representing typical LSA-II material and the experimental programme (set-up of small scale impaction experiments and large scale drop tests, presentation of results) are described. Finally conclusions are drawn.

2 SELECTION OF APPROPRIATE MATERIAL SYSTEMS

Dispersible radioactive material are mainly associated with
- Uranium oxide (UO₂, U₃O₈) in powder form which is employed in enrichment facilities, fabrication of fuel elements and reprocessing plants,
- ashes arising from the incineration of organic radioactive waste,
- resins from ion exchangers, filter sludges, and filter cartridges from cleaning processes,
- building rubbles and excavated soil from the dismantling of nuclear facilities.

Transports of such materials will mainly occur between installations of the fuel cycle and waste management facilities (waste conditioning plants, interim storage facilities, final waste repositories). In order to characterise the above powdery materials with respect to their material characteristics influencing airborne release behaviour available data were evaluated, e.g. from scientific publications/studies or interviews with operators of plants which generate waste or are dealing with waste conditioning.

The experimental programme of our research project comprised small scale impaction tests with small powder specimens (0.1 kg powder mass) as well as real scale drop tests which were carried out for different sizes of powder specimens, i.e. the powder masses ranged between 0.4 kg (glass container) and 260 kg (real Type-A-certified 200 l-standard drum with attached rolling hoops). For these experiments appropriate surrogate powders were identified which, on the one hand, are representative for typical powdery LSA-II materials and, on the other hand, are easily available and economically priced.
The majority of the experiments was carried out using a type of pulverised fly ash (PFA) with a broad particle size distribution between 1 and 100 µm (AED) and a high dispersion propensity characterised as “dustiness”. The size distribution and dispersion propensity of the PFA used can be considered as being representative for ash-type LSA waste and to be conservative for most other powdery LSA-II materials. In Figure 2.1 the particle size distribution of a typical ash originating from the Studsvik-incineration plant (Sweden) in terms of geometric particle diameter is presented together with the geometric particle size distribution for domestic fuel ash. The corresponding aerodynamic particle diameter distribution for domestic fuel ash corresponds to those from PFA, commercially available quartz sand (Millisil® W12) and granulated blast-furnace slag. As a consequence pulverised fly ash was applied as surrogate powder for ash-type LSA waste.

Fig. 2.1: Cumulative particle size distributions for ashes, quartz sand and granulated blast-furnace slag

As an alternate test aerosol titanium dioxide material was chosen. This material system entirely consisted of particles with AED < 1 µm, but showed much less dispersion propensity and a significant lower release rate due to the greater cohesion forces. The TiO₂ material system is considered to be representative for typical uranium dioxide powders. This may be drawn from Figure 2.2 which presents the aerodynamic particle diameter distributions for UO₂ and TiO₂ powders, respectively. The data for UO₂ from Nirex were taken from [2]. The solid curve for UO₂ from Sutter et al. [4] was extrapolated to small particle diameters with the assumption of a log-normal particle size distribution. Additionally, in Figure 2.2 a particle size distribution for the surrogate powder TiO₂ as specified in [4] is shown together with the very narrow banded TiO₂ particle size distribution which was used in the experiments of this study.
3 EXPERIMENTAL PROCEDURE

In order to investigate the release behaviour of airborne dusts from varying quantities of easily dispersible material upon their impact on unyielding surfaces the influence of energy input, package size, packaging and material properties was experimentally analysed.

The pulverised fly ash was used to study the release of airborne and respirable amounts from dust in "uncontained" amounts of powder between 100 g and 20 kg for drop heights of 3.2 to 22 m. This was achieved by glass containers of various sizes which burst on impact. Thereby a release of "uncontained" dust material upon impact is resulting. The enveloped packages used were 10-l sheet-metal canisters as well as Type-A-certified 200-l drums; these were vertically dropped from heights between 3.2 m and 22 m. Table 3.1 summarises the drop experiments. The specimens applied for the drop tests are shown in Figure 3.1. The drop tests were carried out in co-operation with the Federal Institute for Materials Research and Testing (BAM) applying the large drop tower at the test site in Lehre.
Fig. 3.1: Test specimens for the drop test:
small glass container (4 l), Typ A-certified waste drum (200 l), tin can (10 l).

The test specimens were dropped into a 4×4×3 m³ aerosol chamber with an initially open sliding roof (Figure 3.2, left). After the impact of the specimen on the unyielding surface at the bottom of the chamber the roof was immediately closed. The released airborne material of particle sizes up to about 20 µm was homogeneously distributed within the free volume of the chamber by means of a ventilator. Based on the decrease of the measured aerosol concentration with time the initial concentration and thus the airborne material released due to the impact can be derived.

Fig. 3.2: Aerosol chamber
left: with open sliding roof
right: schematic view of instrumentation

In supplement to the drop tests, test bodies of the 100-g category were horizontally impacted at impact velocities that corresponded to drop heights of 6.4 to 64 m (Table 3.1). The test powders were filled into either small cylindrical pill boxes made of tin (cladded specimens, 73 ml) or small glass containers (uncontained specimen, 30 ml). These small scale experiments were conducted using the test rig developed at Fraunhofer ITEM (Figure 3.3). It consists of a pneumatic gun accelerating test specimens with a maximum diameter of 43 mm to velocities typically ranging up to 100 m/s, a hard impact surface, and an aerodynamic classification unit. In this part of the set-up, all airborne particles with aerodynamic diameters
smaller than 100 µm originating from the impact process are separated from the larger fragments in a vertical elutriator. The entire airborne fraction is subsequently further classified in three size ranges between 20 and 100 µm AED and 5 stages below 20 µm using a combination of centrifugal classifier and cascade impactor. Calibration data and performance characteristics of the test rig are presented in [5].

**Fig. 3.3:** Fraunhofer test rig for small scale experiments on release fractions of powder material after mechanical energy impact.

Due to the horizontal impact direction and the special design of the in-situ size classification unit, the apparatus gives reliable and very reproducible information on the generation of dust particles upon impact of powder material or from fragmentation of (brittle) material and, thus, is well suited to explore controlling mechanisms and parameters.

**Tab. 3.1:** Test matrix of the drop experiments (columns 2-6) and the impact experiments (gray shaded cells in row 3)

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4 RESULTS

The uncontained samples (in glass containers) filled with either pulverised fly ash (PFA) or TiO$_2$ behave qualitatively and quantitatively quite different. This can be seen from Figure 4.1. In case of PFA (Figure 4.1, left) the fraction of airborne released material is much higher and consequently the distribution of spilled dust on the ground is much wider and more homogeneous as compared with TiO$_2$ (Figure 4.1 right).

![Pulverised fly ash (PFA)](image1)

![Titanium dioxide](image2)

**Fig. 4.1:** Distribution of spilled material in the aerosol chamber after drop test (drop height: 9 m; volume of glass container: 100 ml).

As a result of the experiments with uncontained samples of the highly dispersive pulverised fly ash, the highest observed airborne release in the respirable particle size range (< 10 µm) amounts to about 2 % for all combinations of dust masses and drop heights from Table 3.1. This maximum release fraction was determined in connection with the drop of 2 kg of uncontained PFA from 9 m height (Figure 4.2). For masses above 1 kg, a trend towards a decrease of the release fraction with increasing mass was observed for uncontained powder masses. For an uncontained dust mass of 100 kg, a respirable release fraction of less than 0.1 % can be extrapolated from Figure 4.2. Up to a drop height of 15 m, the release fraction grows almost linearly with the drop height (Figure 4.3).

**Fig. 4.2:** Release fraction as a function of mass (uncontained), drop height 9m

**Fig. 4.3:** Release fraction as a function of drop height for pulverised fly ash
By enveloping the powder mass in a sheet metal container the release fraction is reduced by 2 – 3 orders of magnitude (Figure 4.3). In the case of the 200-l drum with attached rolling hoops, there was no measurable release up to a drop height of 5.2 m. At a drop height of 9 m, the measured respirable release fraction is $4 \cdot 10^{-5}$. As can be seen from Figure 4.3, the release fraction increases almost proportionally with increasing drop heights up to 15 m. At greater drop heights, the increase of the release fraction is disproportionally low. The extent of the damage to the 200-l drum is limited even if it is dropped from 22 m. The dust is mainly released upon impact onto the lid during the compression and unloading of the cover seal.

The release fractions determined experimentally in the research project of quasi-uncontained fly ash are consistent with the measuring results of other documented studies. In the experiments performed now, the studied range of masses and drop heights was extended considerably towards the upper end. The results that were obtained are therefore much more reliable for real-scale conditions. There has also been a demonstration of the high retaining capacity of a containment corresponding to a higher-quality industry-grade package or a Type-A package under mechanical loads that effectively cover the complete spectrum of possible impact velocities that may occur in transport and handling accidents. The results of this project lead to an improved basis for the determination of the consequences of transport and handling accidents.

5 CONCLUSIONS

For transport or handling accidents involving packages with radioactive materials and the assessment of potential radiological consequences, for the review of current requirements of the Transport Regulations, and for their possible further development reliable release data following mechanical impact are required. Within this context investigations were carried out in order to improve the general physical understanding of the release process as well as the database for the quantity and the characteristics of an airborne release for non-fixed dispersible LSA-II material upon mechanical loading.

Drop experiments applying medium and real sized specimens of different dispersible material (pulverised fly ash and TiO$_2$) have shown that the release behaviour of dispersible powders strongly depends upon material properties, e.g. particle size distribution and cohesion forces. The very fine-grained TiO$_2$ powder (AED $\leq$ 1 $\mu$m) tends to form agglomerates which in turn give rise to an effectively reduced airborne release of the respirable particle fraction (AED < 10 $\mu$m). On the contrary, pulverised fly ash (PFA) with a broad particle size distribution (1 $\mu$m < AED < 100 $\mu$m) showed much higher dispersion propensity and a clearly higher release fraction due to smaller cohesion forces. The highest experimentally determined release fraction of respirable mass (AED < 10 $\mu$m) was obtained for uncontained fly ash and amounts to about 2 % (Figure 4.2). This is, of course, unrealistic because these materials are contained within a packaging when transported.

The application of this respirable release fraction to powdery LSA-II materials following mechanical impact in safety analyses would therefore be overly conservative. For a more realistic quantity of about 100 kg when dropped from 9 m onto a hard surface and no remaining containment is acting upon impact a release fraction of 0.1 % can be assumed. These values may be reduced if the dustiness of the real LSA-II material is known.

Furthermore, the packaging containing the powdery material substantially reduces the release of airborne material. Measured airborne release fractions for a 200 l drum with Type A certificate containing easily dispersible pulverised fuel ash (PFA) were about a factor of 50 to 100 lower than for uncontained material (Figure 4.3). For a drop height of 9 m the airborne release fraction of a 200 l-standard drum amounted to about $4 \cdot 10^{-5}$. This value
should be valid for the majority of transport and handling accidents with mechanical impact. For a metal container of Type IP-2 or better which contains powder masses of 100 kg or more the release fraction of $4 \times 10^{-5}$ after a 9 m drop may be adopted.

Concerning LSA material the current requirements of the IAEA Transport Regulations are based on a simplistic radiological model (often quoted as the 10 mg inhalation model) where it is assumed to be unlikely that a person in the vicinity of an accident site inhales more than 10 mg of released material. LSA-II material is characterised by a specific activity limit of $10^{-4} \text{A}_2/\text{g}$. An intake of 10 mg of such material equates therefore to an intake of $10^{-6} \text{A}_2$. $\text{A}_2$ is the radionuclide-specific activity limit for non special form material in Type A packages and is derived by the method of the so-called Q-system. To the extent that the inhalation pathway determines the $\text{A}_2$ value an intake of $10^{-6} \text{A}_2$ is associated with an effective dose of 50 mSv which is the underlying dose limit of the International Transport Regulations for an individual being exposed in the vicinity of the location of a transport or handling accident.

Based on the experimentally determined release fractions for packages containing LSA-II material in powder form, presented here, and atmospheric dispersion modelling it can be concluded that the resulting inhalation dose of an individual in the vicinity of the accident location lies by more than one order of magnitude below the effective dose limit of 50 mSv. The limits specified in the Transport Regulations of the mass-related activity concentrations for LSA II of $10^{-4} \text{A}_2/\text{g}$ can thus be confirmed as being conservative also for powdery, easily dispersible materials.

6 REFERENCES


