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TESTING OF PACKAGES WITH LSA MATERIALS IN VERY SEVERE MECHANICAL IMPACT CONDITIONS WITH MEASUREMENT OF AIRBORNE RELEASE

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ABSTRACT

To assess the risks associated with transport accidents involving solid LSA-II and LSA-III materials contained in industrial packages a comprehensive experimental program was conducted under the project leadership of GRS to quantify and characterize airborne release of radioactive particulate matter in transport and handling accidents with mechanical impact of varying severities and to determine the dependency from influencing parameters such as LSA material and packaging properties and size. The experimental approach combined well-controlled and very reproducible impact experiments with small scale specimens and drop tests of large scale specimens from different heights up to 27 m. In both cases the associated airborne release of particulate matter was determined by quantifying the amount and aerodynamic particle size characteristics of released dust. In the drop tests the volumes of specimens were varied systematically from 1 to 220 l and drop heights between 5 m and 27 m in order to enable extrapolation to other configurations of package sizes and impact severities. The LSA surrogate materials were either concrete used to immobilize radioactive wastes as representative brittle material or appropriately chosen powders representing dispersible materials. Drop tests were performed with the LSA material either contained within a packaging or without protecting packaging upon impact to determine the influence of the packaging on the airborne release.

Based on the experimental results it can be concluded that the requirements of the current IAEA Transport Regulations sufficiently limit potential radiological consequences from transport accidents with mechanical impact involving packages with LSA-II or LSA-III materials.

INTRODUCTION

For packages with LSA materials a comprehensive experimental program has been conducted to quantify and characterize the airborne release of radioactive particulate in transport and handling accidents with mechanical impact of various severities. The aim of the systematic experimental approach was to establish release data which are applicable to LSA-II and LSA-III materials in industrial IP-2 and IP-3 packages and which allow making predictions about airborne release fractions in relation to mechanical impact severity, mass, and characteristics of LSA material and influence of packaging.

The IAEA Transport Regulations [1, 2] require that packages containing LSA-II or LSA-III materials fulfill a few test requirements, mainly a drop test onto a specified hard and flat target from heights of a minimum of 0.3 m for very heavy packages up to a maximum of 1.2 m for packages with a mass < 5000 kg. These drop tests should essentially guarantee that under normal conditions of transport there is no loss of material from the package and no significant increase of external dose rate.

With respect to accident conditions of transport it is implicitly postulated that a sufficient safety level results from the material requirements and limitations for the allowed specific activity of the radioactive material. In essence, the activity concentration limit of $10^{-4} \times A_2$ / g for LSA-II material is based on the assumption that in case of a (severe) accident, either with mechanical impact or thermal impact from a fire a person in the vicinity of the accident site would inhale no more than 10 mg of airborne released radioactive dust with an activity concentration equal to the $10^{-4} \times A_2$ / g limit. In connection with the modeling approach of the Q-system this implies that in case of an accident the effective dose from inhalation of such material is at most 50 mSv. For LSA-III materials it is implicitly assumed that the amount of airborne released respirable material is lower by at least a factor of 20 and consequently the specific activity of the radioactive material is limited to $2 \cdot 10^{-3} \times A_2$ / g.

The results of the experimental program are applied to estimate the safety margins of the requirements of the current IAEA Transport Regulations concerning potential radiological consequences from severe transport accidents with mechanical impact involving packages with LSA-II or LSA-III materials.

GENERAL INFORMATION ON EXPERIMENTAL PROGRAMME

The main features of the experimental approach were the combination of small scale impact experiments for a multitude of brittle materials revealing fundamental relationships concerning fractional amount and particle size distribution of airborne release as function of impact severity with large scale drop experiments of packaged and unpackaged LSA surrogate materials with drop heights from 5 m to 27 m and measurement of airborne release. A considerable more detailed description of the experimental program, its main results on airborne release and conclusions concerning potential radiological consequences and the adequacy of the respective requirements of the IAEA Transport Regulations has been published very recently [3].

With respect to the performance of packages in accident conditions with mechanical impact the IAEA Transport Regulations define the drop test of a Type B package from a height of 9 m onto an unyielding target as being representative for a very severe accident and many assessments have concluded that thereby a large fraction of severe accident environment are covered. Although the IAEA Transport Regulations do not require IP-2 or IP-3 package containing LSA-II or LSA-III materials to withstand such loads a mechanical impact from 9 m onto an unyielding target is adopted here as reference for a very severe accident. In addition, the test program included drops from 5 m up to 27 m onto an unyielding target in order to determine the dependency of airborne release from impact severity.

A few terms and quantities need to be defined beforehand:

A release fraction, η_x, is defined as the mass, m_x, of released airborne particles with diameter smaller than x normalized to the total mass, M_s, of the material which is subject to energy load.

Particle motion in air, inhalability and lung deposition as well as atmospheric dispersion are all determined by the particle's aerodynamic equivalent diameter, x_{AED}. Therefore, it is mainly used as the particle size parameter, x, in this paper. The aerodynamic diameter (AED) of a particle with arbitrary shape and material bulk density is equivalent to the diameter of a unit density sphere with the same settling velocity in air.

For the reasons mentioned above, special attention is directed to the determination of the airborne release fraction, i.e. particles $x_{AED} \leq 100$ µm, η_{100}, and the respirable release fraction, η_{10} ($x_{AED} \leq 10$ µm). Generally, the values for both release fractions are small: $\eta_{100} << 1$, $\eta_{10} << 1$. Mechanical impact severity defined as specific energy input into a specimen is expressed as specific energy input W_m [J/kg] into a tested specimen of mass M_s defined as the mechanical energy absorbed by the specimen normalized to its mass. In case of an impact of a specimen of mass M_s with a speed ν onto a hard (unyielding) target the kinetic energy of the impacting body is $E_{kin} = ½·M_s·v^2$ and accordingly $W_m = ½·v^2$.
When a specimen is dropped from a height H onto a hard (unyielding) target the kinetic energy at impact is $E_{kin} = M_s·g·H$ (g being the gravitational acceleration) and the specific energy input $W_m = g·H$.

SMALL SCALE TESTS

For the small scale tests an impact and aerosol classification apparatus was applied. Defined energy load is achieved by horizontally accelerating a test specimen and impacting it against a hard target. The pneumatic acceleration device, the dynamic aerosol collection and classification unit and the main results and relationships concerning airborne release have been described elsewhere [3, 4, 5]. The three most important results are:

- For small specimen sizes, and horizontal impact conditions the fractional airborne release η_{100} is linearly dependent of the specific energy input W_m above a certain material dependent (damage) threshold: $\eta_{100} = A + B·W_m$.
- The cumulative particle size distributions for particle sizes in the range ~ 0.1 µm to 100 µm all show a linear dependence of particle size.
- The various brittle materials which were investigated, including different types of cement with a spectrum of typical specifications and immobilized simulate waste materials all show very similar behavior as regards W_m dependence (summarized in Tab. 1) and particle size dependency of fractional airborne release.

Table 1. Regression parameters of the the relationship $\eta_{100} = A + B·W_m$

Material	A [-]	B [kg/J]	ρ [g/cm³]
Glass	3.8E-03	5.2E-06	2.2
RSiC	14.9E-03	4.7E-06	2.5
DUO_2	2.8E-03	6.1E-06	11.0
AlSi	9.3E-04	4.9E-06	1.6
CeO_2	2.8E-03	3.6E-06	6.8
$WZrO_2$	1.9E-03	4.7E-06	10.3
Cement/Concrete waste matrix	3.0E-04	3.0E-06	~ 2.2

It can be seen from Table 1 that the various brittle materials that were investigated in the small scale experiments show a remarkable similarity in the slope B of the dependence on specific energy input W_m, the observed variations lying within about a factor of 2.

LARGE SCALE EXPERIMENTS

The impact tests with measurement of airborne release were performed by dropping specimens of larger masses from a defined height and with pre-chosen orientation onto an unyielding target. Drop heights ranged from 5 m to 27 m including drops from 9 m. The target area was surrounded by a control volume of rectangular dimensions (4 m × 4 m × 3 m) with a roof that was

immediately closed after a drop. The volumes of specimens were systematically increased up to 220 l drums to determine the dependence of airborne release from material size/mass. Drops were performed with the bare material unprotected by a surrounding packaging (termed: unclad) as well as contained within a packaging (termed: clad) to determine in this way the release behavior of the unprotected, bare material and the mitigating influence of the packaging on the airborne release.
Based on the small scale tests with brittle material and taking into account that an important part of LSA-type waste materials are immobilized within a cement/concrete matrix material as representative brittle material a cement/concrete formulation as applied for radioactive waste conditioning was used. For the tests with dispersible LSA-type materials two different powder types were investigated: Very dispersible fly ash powder (pulverized fuel ash, PFA), a large fraction of which being in the respirable size range, and a very fine TiO_2 powder with sub-micron particle sizes but proving to be of much lower dustiness compared to PFA powder.
The packagings used in the drop test were sheet steel metal drums of IP-2 standard in the case of brittle material and with IP-2/Type A certificate for the dispersible materials (Fig. 1). In order to also measure the airborne release from dispersible material following a drop from a given height but without the retention effects of a surrounding packaging the powder was contained within a large glass bulb (Fig. 1) which fractured and disintegrated instantaneously upon impact. Based on the small scale experiments which also included glass as brittle material the contribution of fractured glass particles to the total airborne release could be estimated to be sufficiently low.

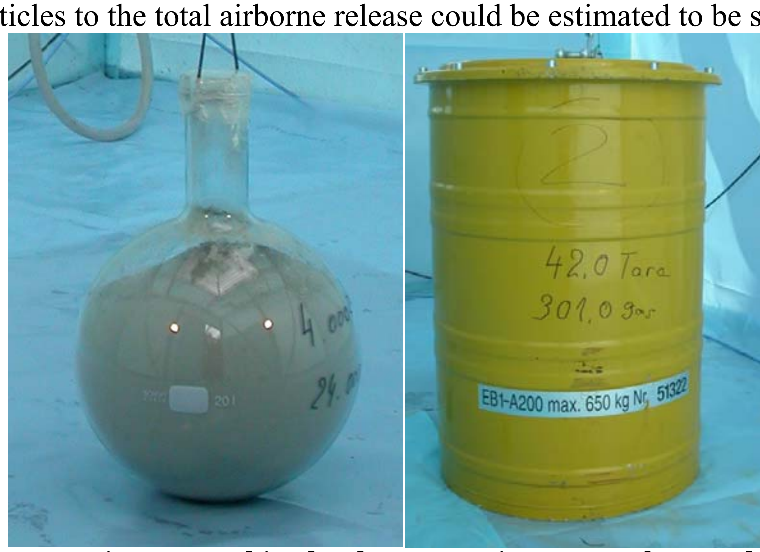

Figure 1. Different specimens used in the drop experiments. Left: powder in a thin 20 l glass flask (unclad); right: 220 l type drum, IP-2/Type A (clad)

In the large scale drop experiments the measurement of airborne particulate release was focused on determination of the respirable particle size range < 10 µm AED: After the specimen impact onto the unyielding target area the subsequent dust release is homogeneously distributed throughout the well stirred (fan) closed control volume. The diagnostics of the release process of airborne material comprises instruments for time and size resolved concentration measurements, cascade impactors and a (high speed) video recording system. The particle concentration decreases exponentially due to particle size dependent wall deposition as well as air exchange between the control volume and the outside environment. Regarding the respirable particle size range the release fraction η_{10} and, hence, the initially released mass below 10 µm AED can be calculated by extrapolating the temporal pattern of the concentration of the integral size fraction $c_{<10}(t)$ to $t = 0$ when impact occurred. Applying the results from the small scale impact experiments for brittle material the airborne release fraction η_{100} for particles below 100 µm AED can be estimated approximately to be a factor of 10 higher than η_{10}.

BASIC RESULTS FROM MEDIUM AND LARGE SCALE RELEASE EXPERIMENTS

Results for brittle material (cement/concrete)

A series of 15 drop experiment with cylindrical specimens made of COVRA concrete simulant were carried out using a test stand operated by Nuclear Research and Consultancy Group (NRG), Arnhem, The Netherlands. These cement/concrete specimens were produced by COVRA, the Central Organisation for Radioactive Waste in The Netherlands according to their formulation used for waste immobilization. The specimens had a volume of 30, 100 and 220 l, respectively and were either clad (9 cases) or unclad. Drop heights were 9 and 27 m. Fig. 2 shows the damage of a clad and unclad specimen (220 l-drums of the same size) falling from 27 m after collection of the debris and partial removement of the sheet steel cladding. The fragment size distribution for the unclad specimen can be qualitatively described as being composed of a few samples with sizes of the order of the test specimen and a large number of fine fragments. All the clad samples have kept their integrity after drop from 9 as well as 27 m. Significant damage occurred to the cement only in the impact zone i.e. the region of maximum stress. Fine particles were released when the metallic skin was damaged preferentially when the specimen impacted on the lid or a weld. A close inspection of the cement after the drum was opened revealed crack patterns on the surface similar to the ones observed for unclad specimen. However, they did not seem to have penetrated the entire specimen since it did not fall apart after removing the entire cladding.

Figure 2. Damage and collected debris of the clad and unclad 220 l-drum after being dropped from a height of 27 m

The measured airborne release fractions with particle sizes below 10 µm (η_{10}) for the 15 drop tests of cement/concrete waste specimens from 9 m (W_m=88 J/kg) and 27 m (W_m=265 J/kg) are summarized in Fig. 3. Measured release fractions for bare (unclad) cement/concrete masses in the range 66 kg to 487 kg are about $2 \cdot 10^{-6}$ in case of a 9 m drop and about a factor of three higher for a 27 m drop. These values are reduced for the 9 m drop height by about a factor of 20 to about $1 \cdot 10^{-7}$ when the cement/concrete specimens are contained in a sheet steel packaging and for a 27 m drop by about a factor of 8 to about 10^{-6}. This observed reduction of the retention effect of the sheet steel container with increasing impact severity is plausible.

When comparing release fractions for (very) small specimens measured with the horizontal impactions apparatus with drop experiments of unclad cement/concrete cylinders covering a range of masses up to 487 kg substantially reduced airborne release fractions < 10 µm are observed with increasing mass/volume. The following approximate empirical relationship for the release fraction for larger masses of unpackaged brittle materials could be derived:

$\eta_{10} = 3 \cdot 10^{-7} \times$ energy input into material mass / (material mass)$^{1.43}$

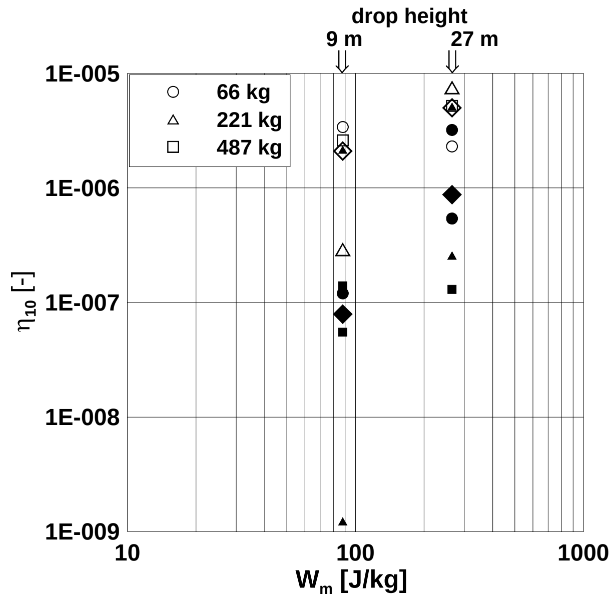

Figure 3. Release fraction η_{10} (AED<10µm) of clad (full symbols) and unclad (open symbols) specimens.
Rhombuses represent the average over the experiments performed at the respective specific energy input

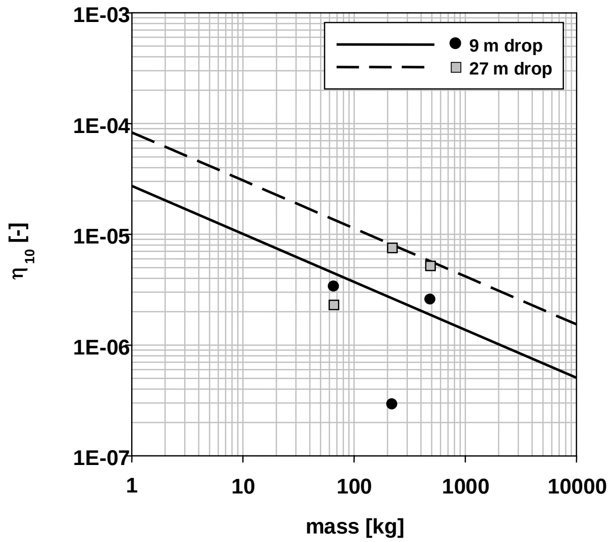

Figure 4. Release fraction η_{10} of unclad COVRA cement "drums" for drops from 9 m and 27 m height onto an unyielding surface as function of specimen mass

This is shown in Fig. 4 for two values of the specific energy input. It should be emphasized that the above relation is based on too few experiments to sufficiently establish a valid relationship for a large range of masses and energy input.

Results for dispersible material

In the drop experiments with dispersible material after a careful choice two quite different types of powders were used: pulverized fuel ash (PFA) which is representative for powders of a high dustiness, i.e. very easily becoming airborne when handled, and a very fine TiO_2 powder with originally submicron particle sizes but of very much lower dustiness because of its sticky nature and therefore tendency to agglomerate. The powders were directly filled into a container – either a glass flask which immediately disintegrated upon impact so that the airborne release was mainly determined by the powder properties or an IP-2 package of 220 l volume and with additional Type A certification. Drop heights were 5 m, 9 m, 15 m and 22 m and with the 220 l package oriented to impact onto the lid edge.

In Fig. 5 measured airborne release fractions with particle sizes < 10 µm AED are shown comparing the drops of a 10 l glass flask filled with 10 kg of the highly dispersible fly ash, representative of unclad material, and of a 220 l drum filled with 260 kg of this powder. In case of the powdery material being uncontained upon impact release fractions are in the range of 10^{-2} and about two orders of magnitude lower (10^{-4}) when contained in a 220 l sheet steel drum. It must be stated at this point that two different masses (10 kg versus 260 kg) were compared here. The difference in release between unclad and clad samples is expected to decrease when equal masses would have been compared. For drop heights < 15 m, the release fraction increases linearly with drop height. For a drop height of 22 m no further increase in release fraction was observed, presumably due to the higher impact speed pushing the dust radially outwards against the walls of the control volume where parts of the dust otherwise released into the airborne state is deposited. A substantial difference in measured airborne release fractions was observed when comparing 9 m drops of glass flasks filled with fly ash or with TiO2 powder, the latter being about 3 orders of magnitude lower.

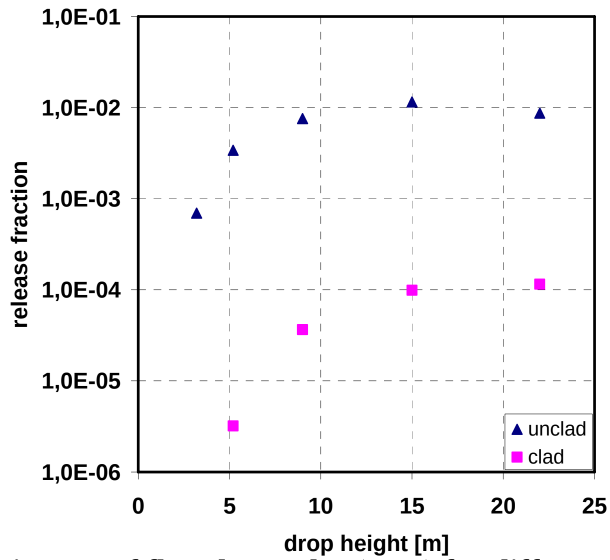

Figure 5. Release fraction η_{10} of fly ash powder (PFA) for different drop heights unclad: 10 kg in glass flask; clad: ca. 260 kg in IP-2/Type A package

CONCLUSIONS ON TRANSPORT SAFETY LEVEL

The results of the experiments for a range of drop heights including a 9 m drop with specimens of representative brittle and dispersible LSA materials, being either unprotected by a packaging or within a sheet steel container up to 220 l volume, can be applied to estimate potential radiological consequences from airborne release and to draw conclusions on the adequacy of the current requirements of the IAEA Transport Regulations. Regarding LSA-II material very unfavorable conditions would be if an IP package filled with highly dispersible radioactive powder, such as ashes, were subjected to a mechanical handling or transport accident with impact severity equivalent to a 9 m drop onto an unyielding target. In this case an airborne respirable release fraction of $4 \cdot 10^{-5}$ could be assumed according to the experimental findings. For a 220 l drum containing a powder mass of 250 kg accordingly an absolute powder release in the respirable size range < 10 µm of $2.5 \cdot 10^{5}$ g × $4 \cdot 10^{-5}$ = 10 g would be an adequate assumption.

Based on considerations of dimensions of the initial dust cloud resulting form the kinematics of an accidental release and conservatively modeled atmospheric dispersion [4] it was concluded that a time-integrated air concentration of $\chi = 10^{-2}$ s/m^3 can be cautiously adopted for an individual being several tens of meters in downwind direction from the accident site during the passage of the released dust cloud. In combination with a breathing rate of an adult of ~$3 \cdot 10^{-4}$ m³ / s this would lead to an intake fraction of $3 \cdot 10^{-6}$ of the airborne release. For the above example of a 10 g particulate release < 10 µm this is equivalent to an intake of 30 µg and therefore well below 10 mg as adopted by the IAEA Regulations for the derivation of the $10^{-4} \times A_2$ / g limit for LSA-II materials. It is immediately evident that also for a much larger package and conservatively assuming the identical release fraction of $4 \cdot 10^{-5}$ there remain substantial safety reserves.

Regarding brittle LSA-type materials which could in principle be at the specific activity limit of $2 \cdot 10^{-3} \times$A2 / g as an example an IP-2 package containing a mass of 2000 kg of e.g. radioactive waste within a cement/concrete matrix is considered here. Based on the 9 m drop experiments a release fraction $\eta_{10} = 10^{-6}$ is derived in this case but with the additional reserve that this value is based on drops of brittle material unprotected by a packaging. This is equivalent to an absolute airborne release of $2 \cdot 10^{6}$ g × 10^{-6} = 2 g respirable material.

With the same line of argumentation as for dispersible LSA-II a person in the vicinity of an accident site and a few tens of meters in downwind direction could inhale a fraction of $3 \cdot 10^{-6}$ of this released radioactive dust leading to an intake of 6 µg. Again this is much lower than the implicit assumption for LSA-III materials that no person in the vicinity of an accident site would have an intake via inhalation of more than 500 µg dust at the specific activity limit of $2 \cdot 10^{-3} \times A_2$ / g.

In summary, it can therefore be concluded that the requirements of the current IAEA Transport Regulations are more than adequate to sufficiently limit potential radiological consequences of accidents as severe as a 9 m drop onto an unyielding target for LSA-II and LSA-III material in an IP-2 or IP-3 package.

This last statement does not mean that the current system of requirements for LSA/SCO materials of the IAEA transport regulations [1] is to be judged as fully adequate. There are, for instance deficiencies in the specification of some of the material requirements for LSA/SCO material which can cause some difficulties for operators in complying with the regulations and to competent authorities in their compliance assurance role. These problems are discussed and a system of revised requirements is suggested in a recent paper [6] which also refers to some of the presented experimental results.

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