Determination of Accident Related Release Data

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ABSTRACT

For accident safety analyses, for the assessment of potential radiological consequences, for the review of current requirements of the Transport Regulations and for their possible further development as well as for the demonstration that radioactive materials such as LDM candidate material fulfil the regulatory requirements 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. In this context special attention has to be paid to radioactive wastes immobilised in brittle materials, e.g. cement/concrete, glass, ceramics or other brittle materials such as fresh and spent fuel.

In this presentation we report on a long-term experimental program 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 siginifcant progress was achieved to meet this objective. The laboratory equipment enables 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 cover 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 exactly measure the amount and aerodynamic size distribution of the airborne release and to quantify its dependence on relevant parameters such as energy input, material properties, and specimen geometry. The experimental program was performed within the scope of various national and international (e.g. EU-funded) 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.

INTRODUCTION

Transported radioactive matter is frequently immobilised in brittle materials, e.g. cement/concrete, glass, ceramics, or is a brittle material by nature such as fresh and spent fuel. International regulations on requirements for the transport of solid radioactive material are based among others on the potential release of aerosol-borne radioactivity under accident conditions. These conditions are in most cases characterized by defined mechanical loadings applied to the system where the radioactive content is likely to fracture and, eventually, to partly disintegrate into aerosol particles. The quantification of the airborne release is an issue of ongoing interest. In order to comply with the demands of radiological consequence analysis the release data should represent the total amount of radioactivity released as airborne particles and the aerodynamic activity size distribution. In this context the airborne size fraction can be defined as all particles having an aerodynamic diameter, AED, smaller than 100 µm. This is of the order of the upper particle size range to be transported in the atmosphere and, thus, posing a risk in view of contamination of ground and other surfaces and inhalation. A subfraction important for exposure assessment is the fraction < 10 µm AED. These particles deposit in the lung and can cause long term radiation burden. The determination of release data has been subject to numerous theoretical and experimental investigations in the past. Many of the experimental studies applied Pellini hammer drop tests on material samples. The fragments were analyzed mainly by sieving after being recovered from the impact apparatus by brushing or rinsing. The drawbacks of nearly all approaches are:

- The methods do not simulate the realistic impact and release process;
- Small respirable particles are likely not to be recovered;

- The particle size is not measured adequately.

In this paper we report on an innovative and versatile approach and an experimental program to characterize fine particle formation and release upon fragmentation of brittle materials after transient mechanical energy input. Our method goes beyond the studies reported on in the literature in various respects: the experimental set-up used for energy input, the method used for characterizing the release, and the particle size range covered.

The rationale behind the program is to carry out well controlled laboratory experiments with small scale non-radioactive brittle test objects in order to reveal the influence of various parameters such as energy input, material properties, specimen geometry etc. on the quantities representing the airborne release, to establish fundamental relationships governing the fragmentation behaviour of various materials and, thus, to improve the general physical understanding of fine particle formation associated with fragmentation of brittle material. The results of the small scale experiments with non-radioactive materials are expected to be valuable for:

- Drawing direct conclusions on source term data for relevant accident scenarios;
- Qualifying possible surrogate materials for real high activity matter such as spent fuel;
- Designing and carrying out a few key real scale experiments with representative package sizes;
- Being used in computer codes on stress distribution for the purpose of extrapolation of the release data to realistic packages based on numerical calculations.

LABORATORY SET-UP

The basic quantity characterizing the release is the cumulative mass (activity) fraction as a function of the aerodynamic diameter, x_{AED}

$$Q_3(x_{AED}) = \frac{m(x_{AED})}{m_s}, \qquad 0 < Q_3(x_{AED}) < 1,$$

where $m(x_{AED})$ is the total mass (activity) of all particles released in the size range smaller than x_{AED}, and m_s is the mass of the test specimen. The values of this function at two special diameters:

$$\eta_{100} = Q_3(100\ \mu\mathrm{m}), \quad \eta_{10} = Q_3(10\ \mu\mathrm{m})$$

represent the total airborne release fraction smaller than 100 µm and the fraction being inhalable into the lung as defined above. In general:

$$\eta_{100} << 1, \quad \eta_{10} << 1.$$

For the small scale tests we developed an impact and aerosol classification apparatus, as shown in Fig. 1. This set-up allows for

- Establishing transient energy input into test specimens of brittle material. This is achieved by either accelerating a test specimen and impacting it against a hard target or shooting a (high speed) projectile onto a suspended brittle object.
- In-situ separation between airborne particles and non-airborne fragments released during the fragmentation process;
- In-situ aerodynamic size classification of the airborne fraction.

In order to simulate impact velocities relevant for accident scenarios for rail and road transport as well as airplane crashes a pneumatic gun is used for acceleration of test specimen. The energy is suffiicient to accelerate objects of 1000 g to velocities up to 120 m/s. The maximum final speed of small masses is 250 m/s. The test specimen is accelerated in a carrier which is abruptly stopped at the exit of the acceleration tube. The object continues to fly with the attained speed and impacts onto a hard unyielding target plate. The actual impact speed is measured optically. Hypervelocity impacts associated with existing scenarios of sabotage attacks on transport casks with high energy density weapons, for example, can be simulated by replacing the pneumatic air gun by a light gas canon or using small conical shaped charges to generate high speed projectiles.

All the devices described above can be combined with a dynamic aerosol collection and classification unit for analyzing the fragments resulting form the impact process. The aerosol apparatus consists of a vertical elutriation box separating the large debris from the airborne particles by suspending them in an upwards directed homogeneous airflow. The flow velocity of 25 cm/s is maintained which is equivalent to the settling velocity of particles with 100 µm AED. Thus, the airborne fraction of the fragments is lifted upwards with the airflow whereas the larger fragments fall on the bottom plate of the elutriator. The air is entering a coarse particle classifier collecting particles in three size intervals covering the range between 100 and 21 µm. In this unit the airflow performs a 180° bend. Suspended particles will be forced outwards due to settling in the centrifugal force field and, depending on their aerodynamic diameter will deposit in different regions of the inner surface of the outer bend wall. Only particles with aerodynamic diameter less than 21 µm are able to pass the bend and are collected on a back-up filter.

A side stream of this fraction is taken and is further classified by a conventional cascade impactor, for example a 150 lpm Berner impactor covering the particle size range between 0.1 and 10 µm with five stages. In total, the measured aerosol size range extends over three orders of magnitude i.e. from 0.1 to 100 µm AED. For each impact experiment, nine samples have to be evaluated either gravimetricaly or by chemical analysis: three samples of the centrifugal classifier, one back-up filter and 5 deposition foils of the cascade impactor. For details of the apparatus see Mädler, 1999 [1].

Non-airborne fragments (>100 µmAED) collected from the bottom of the vertical elutriator can be analyzed by off-line methods such as sieving or laser diffraction spectrometry so that the complete fragment size distribution is characterized.

Calibration data and performance characteristics of the test rig are presented elsewhere [1]. 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 fragmentation of (brittle) material and, thus, is well suited to explore controlling mechanisms and parameters.

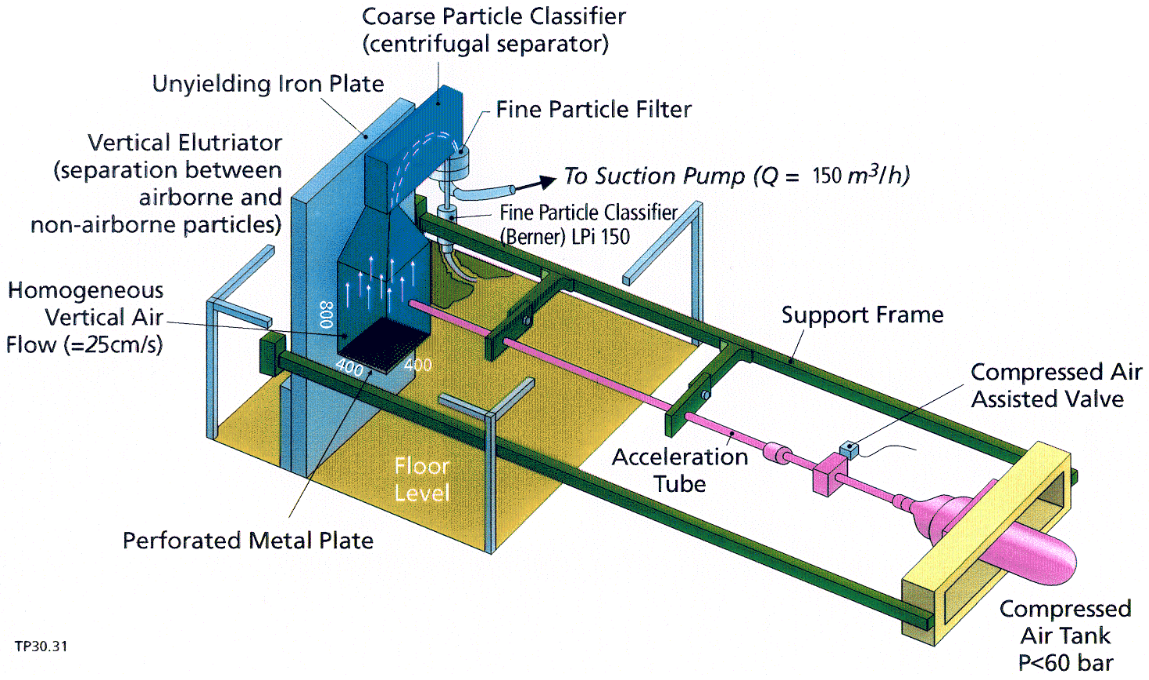

Fig. 1: Laboratory test rig for the characterization of the airborne release of small test specimens of brittle material.

RESULTS

A large number of experiments was performed using the pneumatic acceleration device and impacting test pellets of brittle materials against a hard target. For these experiments the energy input, E, into the pellets is exactly known:

$$E = \frac{1}{2} m_s v^2 \tag{1}$$

where v is the measured impact velocity and m_S is the mass of the test specimen. The well defined experimental boundary conditions and the good reproducibility of the experimental procedure allowed for a systematic study to find out the relevant parameters controlling the formation of the airborne fraction. Plotting the release fraction η_{100} as function of the specific energy input $W_m = E / m_S$ reveals a linear relationship between η_{100} and W_m for W_m-values above a certain material dependent (damage) threshold value:

$$\eta_{100} = A + B\ W_m \tag{2}$$

This is shown in Fig. 2 for small specimens with volumes between 2 cm³ (ceramic materials) and 2-20 cm³ (glass). The ceramic materials - including depleted uranium dioxide (DUO2) pellets - and the glass behave very similar with only small deviations in the slopes obtained from regression analysis performed for each material (see Table 1). Quite obviously there is no correlation between the airborne release fraction and the material density.

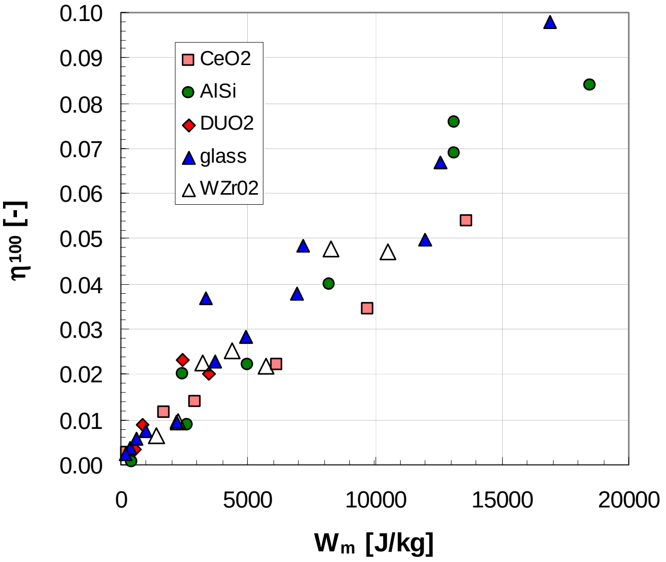

Fig. 2: Release fraction obtained for impact experiments with small cylindrical pellets.

From Fig. 2 it can be derived, for example, that at an impact speed of 100 m/s , equivalent to a specific energy input equivalent to 5000 J/kg, the airborne release fraction of particles below 100 µm is about 3% of the mass of the specimen.

The cumulative mass distribution of the airborne fragments as function of the aerodynamic diameter is described by a universal power law function

$$Q_3(x_{AED}) = a\ x_{AED}^{\mu} \tag{3}$$

as shown for an example in Fig. 3. Here, we plotted the cumulative mass distribution normalized to its value at $x_{AED} = 100$ µm. The shape of the distribution is characterized by a single parameter, the exponent μ. Thus, the measurement of one point of the mass distribution allows for the prediction of the entire aerosol mass distribution in the particle size range between 1 and 100 µm AED. The large number of experiments carried out revealed that the values of the exponent are confined in a relatively narrow range around one. An exponent of μ=1 means a straight line relationship in a linear-linear plot. For the pellet impact tests we did not find any dependency of the scaling exponent, μ, on the energy input and on the type of brittle material tested [2]. Thus, by combining this result with the results shown in Fig. 2 we are able to calculate a very good estimate of the release fraction in any size range smaller than 100 µm AED for a wide class of brittle materials solely on the basis of the specific energy input, W_m. This group of non-radioactive surrogate materials represents relevant nuclear materials such as fresh fuel and vitrified wastes. In view of the mechanically generated release fractions the release data obtained with the above materials provide also a good estimate for spent fuel.

Our results disagree with the correlation suggested in the DOE-Handbook [3]. Here, the respirable release fraction generated mechanically upon impact is proportional to the material density:

$$\eta_{10} = B_{DOE} \ \rho \ W_m \qquad (4)$$

with $B_{DOE} = 2 * 10^{-11} [\mathrm{cm\,s}^2 / g]$ and, thus, results in higher release fractions for DUO_2 than for glass for example. For DUO_2 (ρ = 11 [g/cm³]) at $W_m = 5000$ [J/kg] a release fraction, η_{10}, of 1 % is obtained from the DOE-correlation as opposed to 0.26 % based on Eqn. 2 and Eqn. 3 using the average values of A and B given in Table 1 and μ=1. For glass, both methods result in nearly the same same release fraction (0.2 % versus 0.26%)

Table 1: Regression parameter for the data points of Fig. 2.

Material	A [-]	B [kg/J]	ρ [g/cm³]
Glass	3.77E-03	5.15E-06	2.2
DUO_2	2.82E-03	6.08E-06	11.0
AlSi	9.27E-04	4.93E-06	1.6
CeO_2	2.77E-03	3.56E-06	6.8
$WzrO_2$	1.92 E-03	4.69E-06	10.3

The correlations 2 and 3 do also hold for test specimens made of cement which also showed values of μ close to 1. The regression parameter, B, is approximately a factor 2 smaller than for the ceramic materials. Therefore, the airborne release in relation to the specimen mass following a given specific energy input is about half of that observed for various ceramic materials.

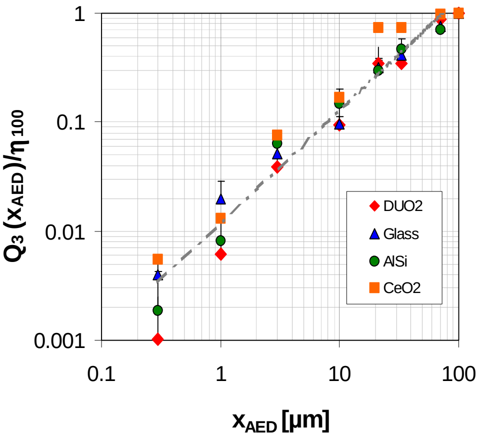

Fig. 3: Cumulative mass size distribution of the airborne release normalized to its value at x_{AED}=100 µm for impact tests with small pellets of various materials (different impact speeds up to 300 m/s). In the submicron size range the scatter of the data becomes quite large for some ceramic materials (DUO2, AlSi) due to the fact that intra-grain boundary cracks are required for the particle formation.

CONCLUSIONS

We have developed a versatile method to study the formation and the release of airborne particles related to transient mechanical energy input into brittle material. A wide range of impact speeds is covered representing road and rail accidents as well as plane crashes. The small scale experiments 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.

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