

AIRBORNE RELEASE FROM VITRIFIED HIGH LEVEL WASTE DUE TO SABOTAGE

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ABSTRACT

A sabotage attack with a shaped charge to a transport and storage cask containing vitrified high-level waste (HLW) could result in the generation of particulate radioactive matter from fractured glass in the cask interior and subsequently to an airborne release to the environment. This potential airborne source term has to be known for the assessment of radiological consequences. A series of small and real scale experiments on the effect of a conical shaped charge (CSC) attack onto non radioactive but chemically equivalent vitrified HLW mockups was performed under the project leadership of GRS in order to quantify the airborne release. The distribution of total mass released, as well as relevant chemical elements were measured as a function of the aerodynamic particle diameter.

Glass aerosol particle and fragment size distributions from CSC impact onto the chemically doped glass targets show patterns generally obtained from brittle fracture upon transient energy input into brittle matter. They follow scaling laws allowing for the prediction of the release of waste material in various particle size ranges based on the measurement of the emitted total mass. The concentration of volatile elements such as for example cesium is found to be enriched in the respirable size range ($<10 \mu\text{m}$) compared to the bulk material. Apart from fracture the high energy of the metallic CSC jet also generates high temperatures within the penetrated area. As a consequence, isotopes of more volatile elements (e.g. Cs 137) can evaporate and re-condense to the aerosol cloud again. This so-called enhancement effect therefore affects the radiological consequences. The enhancement factors obtained in the experiments were significantly below values reported previously by Battelle. Thus, potential radiological consequences are yet lower than previously assumed.

The results from multi element chemical analysis of the aerosol size samples suggest a revision of the understanding of the mechanisms of the enhancement (or enrichment) process. Fine aerosol generated from the lining material of the CSC upon interaction with the target seems to play a key role in the enhancement process. Results are consistent with findings from an ongoing international spent fuel sabotage test program.

INTRODUCTION

A sabotage attack with a shaped charge to a transport and storage cask containing vitrified high-level waste (HLW) could result in the generation of particulate radioactive matter from fractured glass in the cask interior and subsequently to an airborne release to the environment. This potential airborne source term is a prerequisite for the assessment of radiological consequences. High level vitrified waste materials consist of non-volatile nuclides as well as fission products with high volatility such as cesium compounds, for example.

The formation of airborne non-volatile material is related to brittle fracture of the target material. Apart from fracture the high energy of the metallic CSC jet also generates high temperatures within the penetrated area. As a consequence isotopes of more volatile elements can evaporate and re-condense to the aerosol cloud again. This leads to an enhancement of the content of volatile elements in the fine airborne particles compared to the mass fraction of the same element in the bulk material.

The relevant quantity of interest is the cumulative mass of chemical element e , (isotope) as a function of the aerodynamic equivalent particle diameter, d_{AED} , released upon interaction of the inventory material with the CSC. This quantity is denoted by: $M_e(< d_{AED})$, and is measured by accumulating the released material using a sampling device with an upper particle size cut-off, d_{AED} , and analyzing the sample for the chemical elements present in the target material. For quantitative analysis a so called enhancement factor is defined for the volatile fission products, ve , as

$$\text{Eq. 1} \quad IEF_{ve} = \frac{R_{AED}}{R_{bulk}} \quad \text{where} \quad R_{AED} = \frac{M_{ve}(< d_{AED})}{M_{nve}(< d_{AED})} \quad \text{and} \quad R_{bulk} = \frac{M_{ve}(bulk)}{M_{nve}(bulk)}$$

M_{ve} , (M_{nve}) is the mass fraction of the volatile elements (elements considered to be non-volatile) with diameter smaller than d_{AED} , in the particle sample, respectively, the corresponding mass fraction in the bulk material.

Experiments with radioactive and non-radioactive small scale samples performed in the 80s at Battelle Columbus, Ohio already allowed a rough estimation of the source term. From these data a conservative estimate of 50 for the respirable range cesium IEF_{Cs} had been derived. However, the experiments had some limitations, e.g. the reduced scale, the scatter of results and the limited resolution of the relevant particle size range. Later HLW surrogate glass experiments at Battelle had a wider size sampling range but only depleted uranium as a tracer [1]. Therefore, a new set of small scale and large scale experiments was initiated in order to increase the accuracy of radiological consequence analyses.

In this paper we report on the results on the Cs enhancement factor obtained in small and real scale experiments carried out with chemically equivalent but non radioactive surrogate material of the HLW material. Special attention is directed to the size fraction $d_{AED} < 10 \mu\text{m}$ as these particles deposit in the lung and can cause long term radiation exposure.

MATERIALS AND METHODS

Basically the distribution of total mass released, as well as relevant chemical elements were measured by chemical analysis of non-volatile elements of the surrogate material as a function of the aerodynamic particle diameter. We used vitrified waste simulant and experimental set-ups allowing for interaction of CSC jets of different calibers with surrogate targets of different shapes and sizes and at the same time a size selective sampling of the released aerosol. The small scale

tests were carried out with explosive charges fabricated at the explosive laboratory at the Fraunhofer Institute for High-Speed Dynamics, Germany. Finally, real scale CSCs and original transport canisters stored in a transport cask mock-up of original scale were used in the experimental program.

Vitrified Waste Simulant

The test samples were fabricated at the Karlsruhe Research Center during the non-radioactive test operation of a vitrification pilot plant for high-level waste. The material was composed of about 84 wt.% of a borosilicate glass matrix. The remaining 16 wt.% consisted of non-radioactive elements or chemically similar elements representing fission products, actinides and corrosion products. The non-volatile components were replaced by Lanthanum (III) oxide and Neodymium (III) oxide with melting point well above 2000 °C (see Table 1). A representative for the volatile compounds is cesium oxide with a melting point of 490 °C. For the small scale tests we used cylindrical samples with a diameter between 14 and 204 mm and a height between 80 and 200 mm. All samples were clad by a stainless steel mantle. For the large scale test original transport canisters ($\varnothing=430$ mm, glass filling level ~ 1100 mm) for high-activity waste were used.

Table 1. Relevant components of the HLW glass simulant.

Compound	Weight percent [%]	Melting temperature [°C]
Glass matrix:		
SiO ₂ , MgO, CaO, Na ₂ O..	84	-
HLW	16	-
Cs ₂ O	0.44	490
La ₂ O ₃	1.82	2000
Nd ₂ O ₃	1.04	2320

Measurement of the particle size distribution

Small scale tests

Small scale particle size distribution measurements were carried out using an apparatus enabling

- In-situ separation between airborne particles and non-airborne fragments.
- In-situ aerodynamic size classification of the airborne fraction.

This apparatus (Fig. 1) consists of a vertical elutriation box separating the large debris ($> 100\mu\text{m}$ AED) from the airborne particles ($<100\ \mu\text{m}$ AED) by suspending them in an upward directed homogeneous airflow. The airborne fraction of the fragments is lifted upwards, and either passes a foam layer which retains fragments in the size range between 10 and $100\ \mu\text{m}$ AED or, when low dust concentrations are expected, passes a centrifugal classifier resolving three size intervals in the range between 100 and $21\ \mu\text{m}$. The particles penetrating through the respective coarse particle collectors are sampled on a back-up filter. A side stream of this fraction is further classified by conventional Berner cascade impactors. In total, the measured aerosol size range extends over three orders of magnitude ranging from 0.01 to $100\ \mu\text{m}$ AED. For details of the apparatus see [2].

The CSC was placed in front of the classification unit inside of a cylindrical blast shield. Simultaneously with ignition of the CSC a shutter closed the entrance hole in order to minimize the disturbance of the air stream inside the vertical elutriator. The CSC jet was captured by a steel stop block flanged to the classification unit.

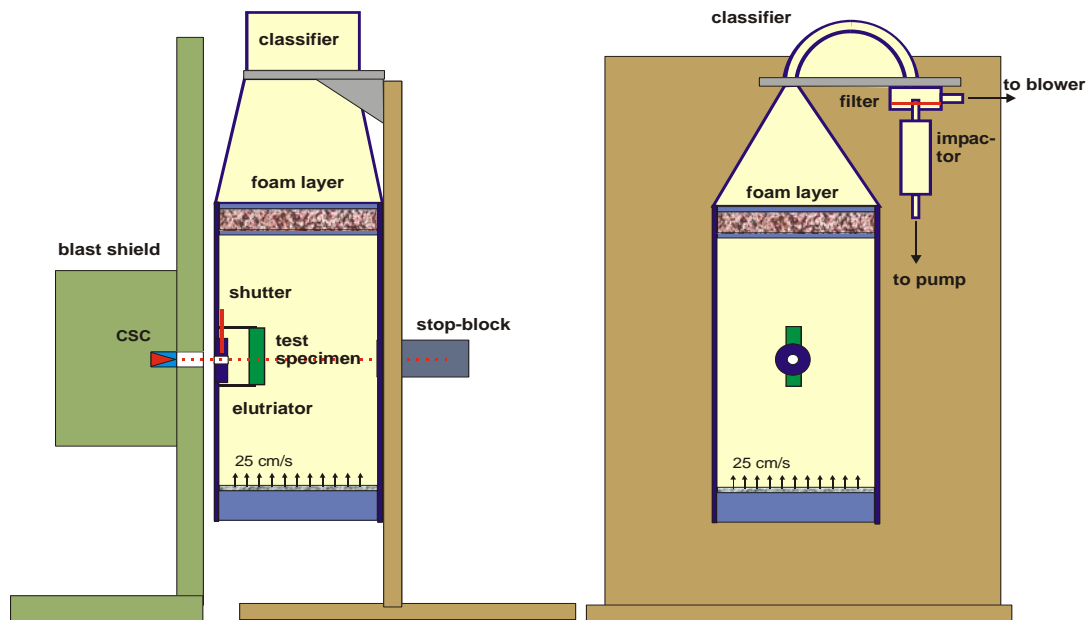


Figure 1: Sketch of the small scale test apparatus.

Large scale tests

The large scale tests were aimed at simulating a realistic accident scenario. Therefore, a flexible transport cask model was designed in order to meet the following requirements:

- Realistic replication with regard to thickness and composition of the cask wall and a realistic free inner volume.
- The possibility to measure the source term in- and outside the cask.
- Compatibility with a real scale CSC.

The transport cask model is an air-tight container made out of reinforced concrete with a wall thickness of 430 mm and a free inner volume of 5.32 m³. The top is sealed with a detachable steel lid. In the front wall a cylindrical steel duct can be equipped with discs made from any desired cask wall material (see Figure 2).

The measuring principle for the aerosol leaving the cask is similar to former tests with depleted uranium [3]. An aerosol sampling chamber is attached to the side wall of the cask where the CSC jet penetrates to collect the total material released out of the penetration channel. The circular jet entrance port is closed by a fast closing slide valve simultaneously with charge ignition. The airborne particles (< 100 μm) are suspended in a vertical elutriator (see above) with an air flow of 220 m³/h. The airflow subsequently passes through a 4-stage coarse particle impactor (100-20 μm, 20-10 μm, 10-5 μm and < 5 μm). In parallel to the main air stream several isokinetic sampling tube performed side stream sampling to additional Berner impactors and a time resolved aerosol spectrometer (Respicon™, [4]) with a size resolution : < 5, 5-10, 10-80 μm AED.

The concentration history was also measured inside the cask an aerosol photometer and a Respicon™. Furthermore, two fast response thermocouples and one fast response pressure gauge were installed. In total five small and four real scale tests with clad specimen were carried out.

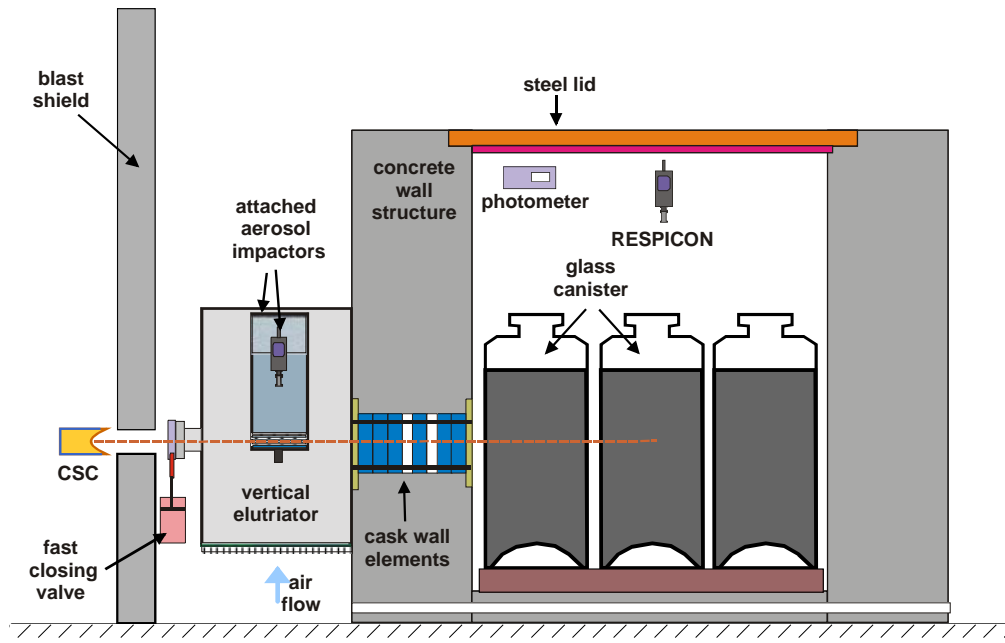


Figure 2: Sketch of the model cask made from reinforced concrete.

RESULTS

Table 2 represents the test matrix and the results obtained for the Cs enhancement factor in the respirable size range. In contrast to the Battelle Columbus results our measurements with HLW simulants reveal considerably lower enhancement factors with a high variability though, ranging from 1.3 to 7.6. Figure 3 shows differential enhancement factors determined from the ratios as defined by Eq.1 for every single size interval resolved by the cascade impactors. It is directly obtained by comparing the Cs size distribution and the La size distribution. An example of the respective elemental size distribution is shown in Figure 4 together with the size distribution of copper and the total aerosol mass.

Table 2: Test matrix and integrated enhancement factor for particles < 10 μm AED (IEF(10)).

Exp. No.	Target	CSC	IEF(10)
s #1	cylinder, \varnothing 14 mm		7.6
s #2	cylinder, \varnothing 42 mm		1.7
s #3	cylinder, \varnothing 42 mm	small scale	2.0
s #4	cylinder, \varnothing 104 mm		2.2
s #5	cylinder \varnothing 204 mm		1.7
l #1	cylinder, \varnothing 104 mm		2.8
l #2	1 full sized canister	full scale	4.4
l #3	3 full sized canister		2.6
l #4	3 full sized canister		1.3

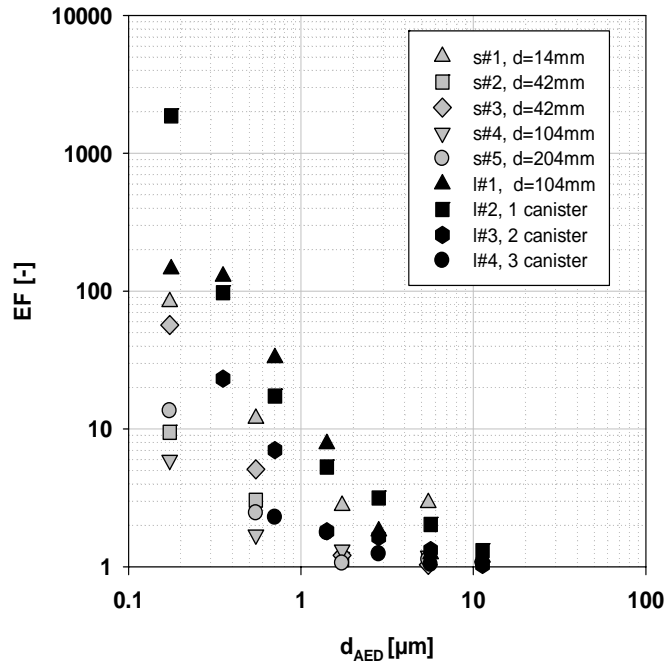


Figure 3: Differential enhancement factors

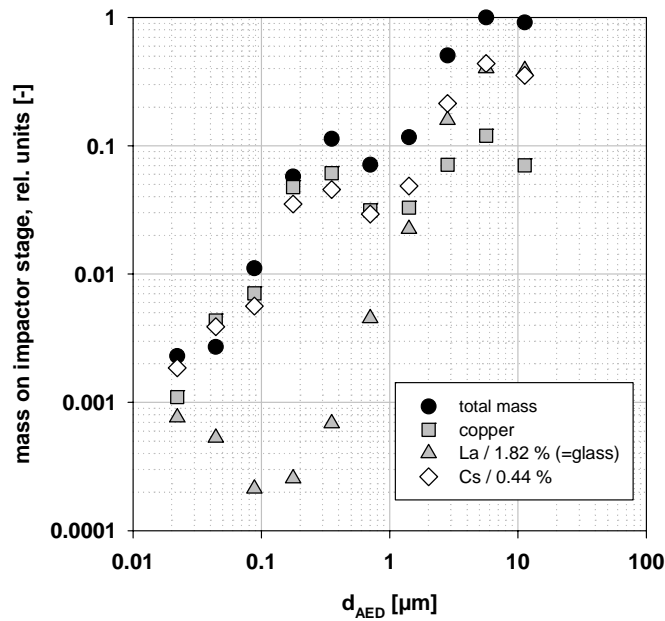


Figure 4: Differential particle size distribution for copper, glass (as determined from the lanthanum analysis), cesium normalized to its content in the bulk glass material (0.44%) and the total mass. (Small scale experiment s #2).

The stage loadings of the chemical elements vary over four orders of magnitude. In the submicron regime the total mass is dominated by the copper whereas above 1 μm the contribution of the glass becomes more and more important. The mass balance is closed by soot from the explosive and iron released from the cladding material, cask wall material (in case of the large scale experiments), respectively the stop block of the jet. There is a significant

difference in the size distribution of the (non-volatile) lanthanum and the elemental size distribution for copper and cesium. The La size distribution is essentially unimodal except for a very small fraction in the size range below 0.1 μm , as opposed to the cesium and the copper distributions which show a pronounced bimodality with a condensation peak at around 0.5 μm . Below 1 μm the Cu and Cs size distributions are remarkably identical: Above 3 μm the Cs as well as the Cu distribution have a shape similar to the shape of the glass distribution.

The detailed particle size resolved multi-element analysis suggests that copper from the hot CSC lining material plays a key role in the release and the re-condensation of the volatile Cs. The process may be regarded as a binary nucleation process, where Cu and Cs vapor generated in the hot interaction zone of the CSC-jet with the target undergo simultaneous particle formation upon cooling. For the particles above 3 μm the ratio between the volatile Cs and the non-volatile La approaches to the ratio in the bulk material. The analysis of Cs and La in the large scale experiments reveals basically the same trends as those of the small scale tests.

We hypothesize that the enhancement factor for the volatile elements of the HLW-simulant as defined above is determined by independent processes:

- The local process of vaporization of volatile material from the target material and comparatively large quantities of copper in the hot interaction zone of the jet and subsequent formation of a copper fume incorporating the volatile elements of the HLW simulant.
- Generation of respirable particles by shock wave induced fragmentation of the glass matrix. This process takes place in a volume of the specimen (swept volume) which is larger than the assumed volume of the hot interaction zone.
- Possible interaction between these particle modes, for example coagulation.

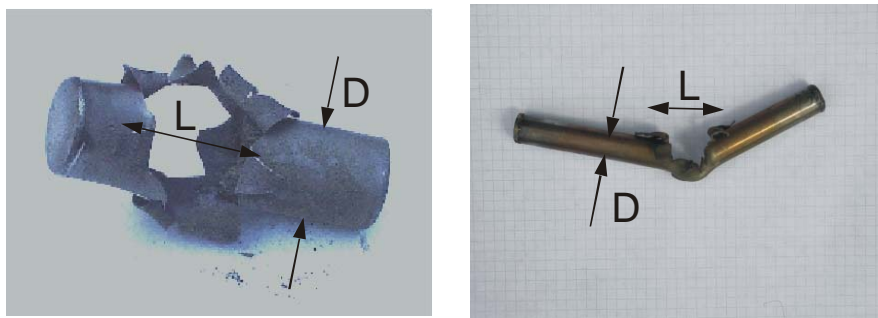


Figure 5: Destroyed sections of the cylinders used in test s #2 (left), and s # 1 (right). The same CSC was used in both cases.

Figure 5 shows two small scale cylinders after impact with the small scale CSC where the dimension of the damage transverse to the jet axis is limited by the cylinder diameter D . The swept volume, V_s , can be clearly identified and quantified: $V_s \propto L D^2$. For the volume, V_i , of immediate interaction between jet and target it is reasonable to assume that it scales with the jet diameter, $d_j \ll D$, and the interaction length, D , in case the jet penetrates through the entire cylinder: $V_i \propto d_j^2 D$. If it furthermore assumed that the respirable mass of lanthanum, $M_{La}(<10\mu\text{m})$, is proportional to the swept volume, V_s , and the respirable mass of the volatile cesium, $M_{Cs}(<10\mu\text{m})$, is proportional to the interaction volume, V_i , the enhancement factor $IEF(10)$ should scale as: $IEF(10) \propto d_j^2 / (LD)$ (see Eq. 1). Since the same CSC was used in both

experiments, a larger enhancement factor is expected in experiments #1 compared to #2. Using the estimated values of L and D given in Figure 5 a factor of 4 between the corresponding $IEF(10)$ values is calculated which is about the number calculated from Table 2. For larger glass specimen relative to the CSC both volumes V_i and V_s are limited by D in jet axis direction only and $IEF(10) \propto d_j^2 / L^2$ which seems to be a universal ratio for all CSC scales. Hence, cesium IEF data of all other experiments are rather similar and independent on the test scale.

CONCLUSIONS

The integrated cesium enhancement factor in the respirable particle size range for vitrified HLW is significantly below the estimated factor derived from former Battelle experiment which only resolved the small diameter part of the respirable aerosol range. These new results are supported by recent comparative glass tests in a different semi-open test apparatus at Sandia National Laboratories within an international sabotage test program on spent fuel release [5]. In our experiments we found no significant differences in the enhancement factor when we scaled up both, the target and the calibre of the CSC as long as the target diameter does not limit the extent of damage.

Detailed particle size resolved multi-element chemical analysis gave some hints on possible mechanism involved in the complex process of the redistribution of volatile elements into the fine particle fraction. Maximum aerosol release data from these experiments are in good agreement with earlier upper estimates based on the small scale Battelle experiments. Due to the reduced enhancement factor potential radiological consequences of a sabotage attack against a HLW transport and storage cask are yet lower as previously assumed.

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