RELEASE OF POWDERED MATERIAL FROM WASTE PACKAGES

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ABSTRACT

Possible incidents in the operational phase of the planned German repository "KONRAD" for radioactive waste with negligible heat production were investigated to assess the radiological consequences. For these investigations, release fractions of the radioactive materials are required. This paper deals with the determination of the release of powdered material from waste packages under mechanical stress. These determinations were based on experiments. The experimental procedure and the process parameters chosen in accordance with the conditions in the planned repository will be described. The significance of the experimental results is discussed with respect to incidents in the planned repository.

INTRODUCTION

Investigations of assumed incidents during the operational phase of a repository for radioactive waste are a precondition for the licensing procedure for the repository in the Federal Republic of Germany. For the assessment of the radiological consequences of such incidents, release fractions of radionuclides have to be determined. In this investigation, several chemical and physical parameters have to be considered. For example, the release fractions depend on the thermal and mechanical stresses on the waste package as a result of the incident. In addition, the release fractions are influenced by the physical and chemical properties of the waste and the waste package. It was thus necessary to determine release fractions on the basis of model assumptions or experimental results. The application of these methods requires the use of conservative or upper limit data for release fractions.

The waste acceptance criteria of the planned German repository "KONRAD" permit unfixed solid waste including powdered material. This is a distinct difference to the acceptance criteria for radioactive wastes in repositories in USA. Since it is not possible to check the amount of powdered material contained in waste packages using the product control, there is no limit in FRG for the particle spectra, as apposed to the US-practice for "Contact Handled"-waste.

A larger amount of powdered material can be expected in waste packages containing incinerated waste (ash) or filter aids. In the planned repository, waste packages will be handled at a height of one to five meters above the ground. If a container drops from these heights, a release of radioactive materials, especially of powdered material, can be expected for certain kinds of waste packages. The radiological consequences of such incidents depend on the size and mass distribution of the released particle spectra. The spectra of the particles released will be determined by the particle spectra of the powdered waste, the height of fall, the mass of waste and the possible ventilation conditions. In order to model the release behavior in a reasonable manner, experimental investigations were carried out. The experimental procedure and the results will be described.

EXPERIMENTS

For the determination of release fractions of powdered material from waste packages under mechanical stress, a test facility was designed to carry out drop tests on containers filled with powdered material. The main component of this facility was a room with walls made of precious metal. The ground area of this room was 3 m by 4 m and the height 3 m. The roof of this test room was equipped with a special chimney with which drop heights of up to 5 meters were possible. Instruments were installed in the room to measure the dependence of the spreading powder cloud after the impact on time, height and particle size. A ventilation, which induced an air stream of 1 m/s at the impact point at a height of 10 cm to 15 cm above ground, was installed. An air circulation providing a complete air exchange within 4 hours was continuously working during the experiments. An illustration of this room (birds eye view) is shown in Fig. 1. With filters of glass-fibre (No. 1, 2 and 3 in Fig. 1), the time-dependence of the spreading cloud was detected. With analog filters (4,5 and 6) the height-dependence was measured. The nuclepore filter (7) was used for taking electronmicroscopic pictures of the largest particles after the impact. An aerodynamic particle sizer (8) for the measurement of the fine powder was installed on a board, with a height of one meter, on the right side of the room. A cup containing a glass shell was placed on the floor (9). Particles which sedimented on this shell were investigated. On the left wall, glass shells were fixed at different heights. Powder, which was deposited on these shells after diffusion and turbulence processes, should enable an assessment of the

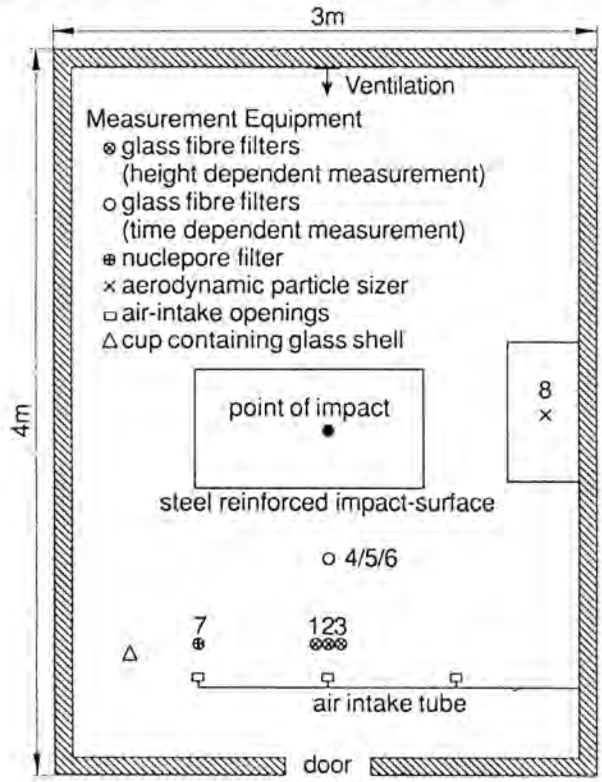

Fig. 1. Test Facility with Installed Measurement Equipment

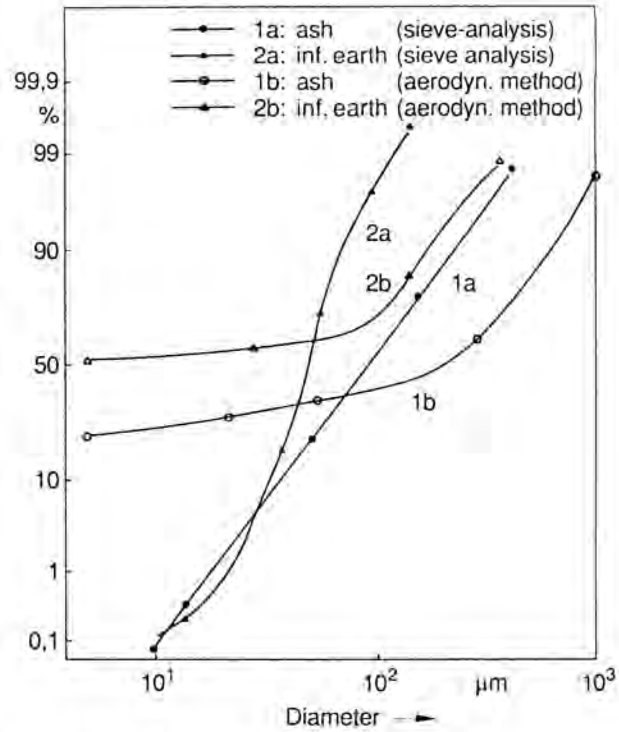

Fig. 2. Cumulative Mass Frequency for Infusorial Earth and Ash.

masses discharged on the wall. With a camera recorder, the drop experiments could be observed.

For the drop tests, a special roofless wooden container was used. This container had the dimensions of 25 cm by 25 cm by 25 cm. The walls of the container had a special construction. They flapped aside instantly at the moment of impact. Thus, any retention effect of the container is neglected. Considering the determination of release fractions for real waste packages, this is a conservative approach.

A sensitive point in the experiments was the powdered material. It was necessary to work with inactive waste. On the other hand, the simulated waste has to be realistic and comparable to the real waste. Hence, the use of two different materials, matching the size-distribution of real waste, was suggested. Experiments with inactive infusorial earth and ash gained by incinerated wood should be carried out. In order to prove their suitability, these two materials were investigated in the preparation of the experiments. The ash was first sieved with a mesh width of 2 mm. Then the size-distributions of the two materials were determined by a sieve-analysis. The results of this analysis are shown in Fig. 2 (curves 1a and 2a). The x-axis in Fig. 2 represents the geometric diameter, the y-axis the relation of mass to the total mass in percentage. For the curves mentioned, the figure shows a mass-relation of only 0.07 % for ash particles and only 0.1 % for infusorial earth with a diameter less than 10 μm. Comparisons of these results with measured fractions of fine powder before testing, yield resulting relative release fractions for diameters below 10 μm of about 100 %, and sometimes even higher values. This result indicated a failure of the sieve-analysis. In a next approach, the size-distribution was redetermined by an aerodynamic measurement (vertical elutriation). The result of this determination is shown in Fig. 2 (curve 1b for ash and 2b for infusorial earth). For these splines, the x-axis represents the aerodynamic diameter. The percentage of fine powder below a diameter of 10 μm is 20 % for ash and 50 % for infusorial earth.

EXPERIMENTAL PROCEDURE

Table I gives an overview on the experiments performed. Twenty drop tests were carried out with heights varying between 1 m and 5 m. Four tests are immediately relevant in the view of incidents during the operational phase of the planned repository.

Exp. 6 : 5m drop height with ventilation

Exp. 20 : 2m drop height with ventilation

Exp. 10 : 3m drop height without ventilation

Exp. 19 : 1m drop height without ventilation

Several tests with different masses and ventilation conditions were performed in order to investigate ventilation

TABLE I
Overview of the Experiments Carried Out and Relevant Process Parameters

Waste	Number of Exp.	m_o (kg)	h (m)	Ventilation
ash	1	6.00	5	no
ash	2	2.00	5	no
ash	3	0.50	5	no
inf. earth	4	0.25	5	no
inf. earth	5	1.00	5	no
inf. earth	6	1.00	5	yes
inf. earth	7	1.00	3	yes
inf. earth	8	1.00	3	yes
inf. earth	9	1.00	3	no
inf. earth	10	1.00	3	no
ash	11	6.00	5	yes
ash	12	0.50	5	yes
ash	13	6.00	3	no
ash	14	6.00	3	yes
inf. earth	15	0.25	5	yes
ash	16	6.00	5	yes
inf. earth	17	0.25	5	yes
inf. earth	18	1.00	1	yes
inf. earth	19	1.00	1	no
inf. earth	20	1.00	2	yes
inf. earth	21	1.00	2	no

and mass effects for one drop height. The mass of ash used was between 0.5 and 6.0 kg and the mass of infusorial earth was between 0.25 and 1.0 kg. The waste container was completely filled with 1 kg of infusorial earth; higher masses could not be taken into account.

In order to record the size-distribution of the spreading powder cloud after a crash of a waste container, the Aerodynamic Particle Sizer (APS, point 8 in Fig. 1) transmite the number of particles/channel online to a microcomputer. The APS has 50 channels which cover a range of aerodynamic diameters extending from less than 0.486 μm to more than 15.4 μm.

The filters of glass-fibre, which were used to determine the dependence of the spreading powder cloud to time and height, were limited to particles with an aerodynamic diameter less than 70 μm. The filters 1, 2 and 3 (see Fig. 1), were successively started with a delay of 10 min each to determine the time dependence. The filters 4, 5 and 6 (see Fig. 1), were installed at heights of 0.4 m, 1.0 m and 1.6 m to determine the height dependence. These filters started measuring immediately after the impact.

EXPERIMENTAL RESULTS

The integral concentration of discharged particles, normalized to the volume of the test room as a function of time, is shown in Fig. 3 for ash and Fig. 4 for infusorial earth. The bar-charts represent the mass-concentration discharged in the room atmosphere, as a function of the height of drop, the mass of waste and different ventilation conditions. At

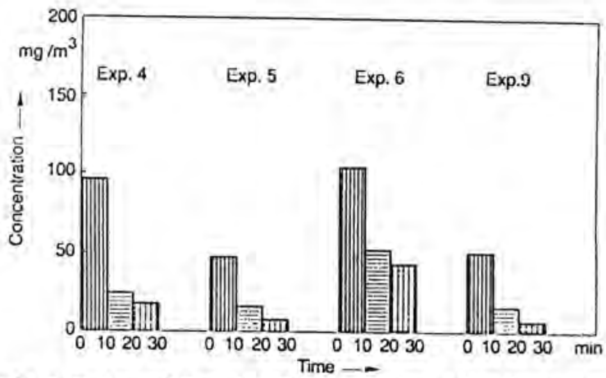

Fig. 3. Released Powder Concentration After Impact as a Function of Time (Ash).

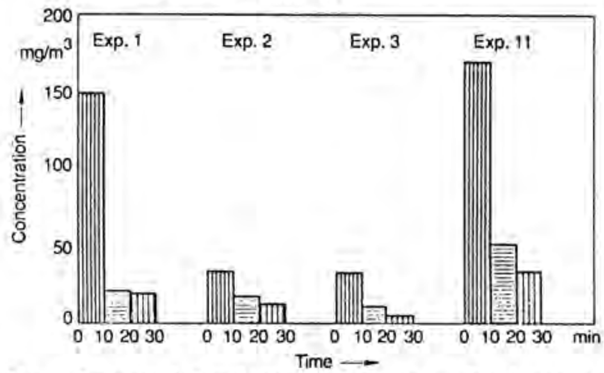

Fig. 4. Released Powder Concentration After Impact as a Function of Time (Infusorial Earth).

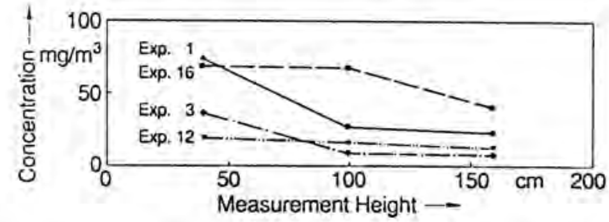

Fig. 5. Released Powder Concentration After Impact as a Function of Height (Ash).

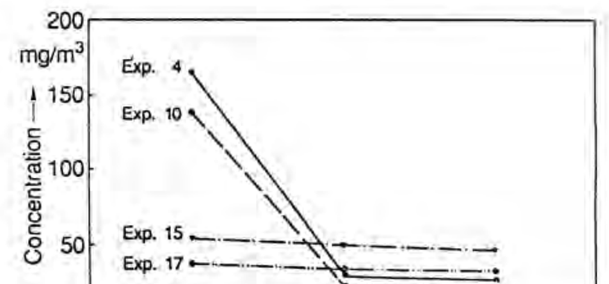

Fig. 6. Released Powder Concentration After Impact as a Function of Height (Infusorial Earth).

first look the bar-charts show a similar behavior, independent of the process parameters and the kind of waste. A decrease of particle concentration with time is generally to be expected. This fact is predominantly caused by the mass of waste. Nevertheless, this effect will be negligible for smaller masses. For smaller masses, the retention effect of the powdered material is negligible. This causes the integral discharged particle quantity to become nearly independent of the mass of waste. By using larger masses, the absolute discharged particle quantity will be increased because of the greater mass supplied.

The discharged particle quantity increases with the ventilation. This effect can be seen by comparing the results of experiments 1 and 11 (see Fig. 3). This is due to the fact that the behavior of infusorial earth release is comparable to the behavior of ash. The mass of the integral discharged particle quantity as a function of the height is shown in Fig. 5 for ash and in Fig. 6 for infusorial earth. The concentration of discharged particles was measured at heights of 0.4 m, 1.0 m and 1.6 m (see Fig. 1 point 4, 5 and 6). For the height of 0.4 m, the integral discharged particle quantity strongly depends on the drop height, the mass of waste and the ventilation conditions. Without ventilation, the reduction of the mass of waste or of the drop height causes a lower integral powder concentration at 0.4 m. In any case, the difference between the measured powder concentration at the heights of 1.0 m and 1.6 m is small. These effects have been observed both in the experiments with ashes as well as with infusorial earth. In experiments with active ventilation, the height dependence was nearly negligible. For all three heights, the concentration of discharged particles measured was almost the same. However, for the comparable experiments with the same process parameters but without ventilation, lower concentrations were measured in the upper levels. This effect can be seen in Fig. 6 for infusorial earth. Although the same effect is obtained for ash, a different behavior is observed in experiment 16 (Fig. 5). This could be caused by experimental uncertainties. The ventilation effect could generally be explained by a retention effect of the air stream. For particles, the air stream can have the effect of a wall. When particles penetrate the airstream, they are uniformly distributed over the entire height .

In Fig. 7, the normalized released particle quantity is shown as a function of waste mass. The lines in this figure link measured experimental ratios for experiments where the mass of waste was the variable process parameter. In Fig. 7, this correlation is shown for ash and infusorial earth for different drop heights.

Considering these facts, the following statements could be made:

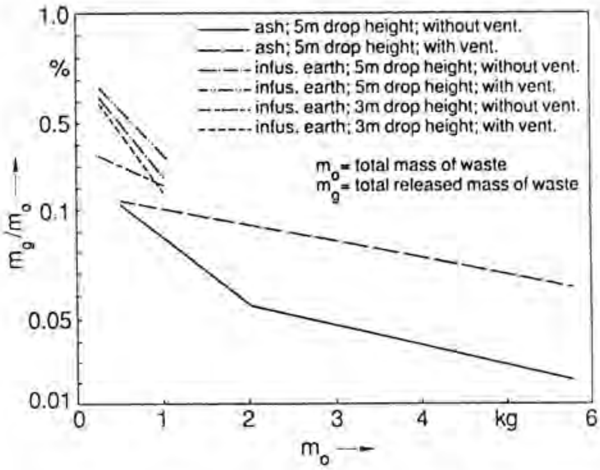

Fig. 7. Relative Release Fractions as a Function of the Total Mass of Waste.

- the relative release fraction decreases with increasing waste mass
- the release fractions increase under ventilation conditions
- the release fractions increase with increasing drop heights
- the release fractions are distinctly higher for infusorial earth than for ashes

By plotting the released normalized fine particle quantity in the same manner as in Fig. 7, the same qualitative results are obtained.

A selection of measured released particle fractions in the experiments with 1 kg infusorial earth are shown in Table II. Here, both absolute and relative release fractions are listed. The first column lists the drop height, the second the released particle mass for fine powder with an aerodynamical diameter of less than 10 μm, the third for the range between 10 μm and 70 μm and the fourth the values for an aerodynamical diameter greater than 70 μm are listend. The

TABLE II

Absolute and Relative Fractions of Released Mass for Selected Experiments.

Drop. Height (m)	m_F (mg)	m_2 (mg)	m_3 (mg)	m_F/m_{OF} (%)	m_2/m_{OF} (%)	m_3/m (%)
5	176.4	3256	69500	0.035	2.83	18.2
3	67.7	2066	20700	0.013	1.80	5.4
1	19.1	2219	2219	0.004	0.86	0.5
2	115.2	9113	9113	0.023	1.31	2.3

next three columns list the ratios of the intervals of release fractions with respect to the source quantity in the corresponding interval. The mass fraction of particles with an aerodynamic diameter less than 10 μm were originally correlated with a separation curve. Such values are needed for conventional calculations. For the determination of radiological consequences, this separation behavior is a part of the dose factors. The originally measured data for the particle fraction below 10 μm diameter were transformed. The transformed data are listed in the column 2 of Table II.

APPLICATION OF EXPERIMENTAL RESULTS

The experiments described above were carried out in order to gain release fractions of radioactive material from waste packages under mechanical stress. The four relevant incidents in the repository, where waste packages can drop from transport equipment and radioactive powder could be released, are:

- drop from a height of 5.00 m under ventilation conditions
- drop from a height of 1.75 m under ventilation conditions
- drop from a height of 3.00 m without ventilation conditions
- drop from a height of 1.20 m without ventilation conditions

The measurements are the basis for the following determination of release fractions caused by the incidents mentioned above:

- Only the results from experiments with infusorial earth were used for generating release fractions.
- As a further constraint only experimental results with 1 kg infusorial earth were taken into account.

This approach is very conservative and is thus assumed to cover experimental uncertainties.

Due to this approach, the experiments 6, 10, 19 and 20 are the basis for further considerations.

For the calculations of potential radiological consequences caused by mechanical stress on the waste package, released particles with an aerodynamic diameter less than 100 μm have to be taken into account. In Fig. 8, the cumulative mass frequency of the released and the supplied powdered material are shown as a function of the aerodynamic particle diameter. The splines are constructed by measured data up to 15.4 μm and at 70.0 μm. The data below 15.4 μm were gained from the measurement with the aerodynamic particle sizer. The value at 70 μm was determined by the filter measurements. In Fig. 8, it can be seen that the step between 15.4 μm and 70.0 μm could be fitted by a linear interpolation in the log-normal representation.

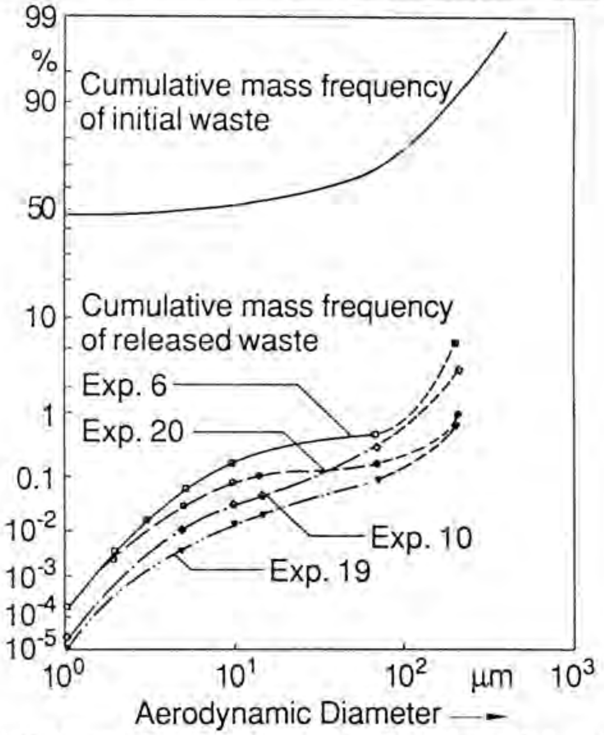

Fig. 8. Cumulative Mass Frequency of Intial and Released Waste.

Due to experimental uncertainties, the range between 70.0 μm and 100 μm could not be fitted in the same manner. This region was extrapolated on the basis of the released particle quantity in the interval 60.0 μm to 70.0 μm. A compilation of measured and calculated release fractions for the four described experiments is shown in Table III. These release fractions were used for the assessment of radiological consequences of incidents with radioactive powder released from waste packages under mechanical stress.

TABLE III
Cumulative Mass Frequency Used for Describing the Radiological Consequences

Aerodynamic Diameter (μm)	Cumulative mass frequency			
	H=5m, V=yes	H=2m, V=yes	H=3m, V=no	H=1m,V=no
5.0E0	5.8E-4	2.7E-4	1.4E-4	4.5E-5
1.0E1	1.7E-3	6.2E-4	3.9E-4	1.1E-4
2.0E1	3.8E-3	1.0E-3	9.5E-4	2.2E-4
3.0E1	4.3E-3	1.1E-3	1.2E-3	3.5E-4
4.0E1	4.7E-3	1.2E-3	1.5E-3	5.0E-4
5.0E1	5.0E-3	1.3E-3	1.7E-3	6.2E-4
6.0E1	5.3E-3	1.4E-3	1.9E-3	7.3E-4
7.0E1	5.6E-3	1.5E-3	2.1E-3	9.2E-4
8.0E1		1.5E-3		
9.0E1		1.6E-3		
1.0E2		1.7E-3		

H = Height; V = Ventilation

The radiological consequences determined, were the basis for the derivation of activity limits for a large number of radionuclides in waste packages with respect to incidents. These limits will be a part of the acceptance criteria for waste in the planned repository "KONRAD".

CONCLUSIONS

Drop experiments with simulated waste were carried out in order to determine release fractions of powdered material from waste packages under mechanical stress. The particle size distribution of the simulated waste was realistically chosen. It was assumed that the waste packages are completely filled with powdered material. The dependence of release fractions on different parameters was investigated. All parameters with the exception of the waste mass were comparable to the conditions in the planned German repository "KONRAD". The experiments have qualitively shown that with increasing waste mass the relative fractions of released particle decreased. Since real waste containers contain several tons of waste, the use of the experimental results to describe the incidents in the repository is a conservative approach. On the basis of the qualitative and quantitative discussion of the experimental results, it was possible to obtain release fractions for the relevant incidents in the repository. Even by taking into account experimental uncertainties, it can be assumed that the release fractions determined on the basis of experimental results, are conservative upper limit values.