ACCIDENTAL RELEASE OF RADIOACTIVITY FROM WASTE PACKAGES IN A FINAL WASTE REPOSITORY

Detlef Gründler

Gesellschaft für Reaktorsicherheit (GRS) mbH
5000 Köln, Federal Republic of Germany

ABSTRACT

The determination of accidental releases of activity is an essential part of any incident analysis. The present paper uses the example of cemented wastes to describe the possible quantification of released activity considering certain given design loads. First, a survey of experimental investigations relating to the release of activity from cement products is presented. Here, the distribution functions of particles are measured which result from mechanical stresses acting on the product. As the results of these experiments are only valid for the prevailing special experimental boundary conditions, it is necessary to solve the transferability problem. For this purpose, a method is presented by which experimentally determined distribution functions can be theoretically converted to other boundary conditions. The method is also suited for a general calculatory determination of the release of activity from cemented waste packages as a function of given design loads.

INTRODUCTION

Within the scope of the licensing procedure for a final repository for radioactive wastes, it has to be demonstrated that the accidental release of radioactivity is so low that it remains below given incident dose limits imposed by the authorities. As far as a final repository is concerned, the only release to be investigated for this demonstration is that from waste packages as a result of the stresses occurring during the incident.

Such an incident analysis is based on plant design. Due to fixed geometric and physical quantities, this design incorporates design loads acting on the waste packages during an incident. The design loads, in turn, determine the extent of the activity release.

The paper deals with the last phase of an incident analysis, i.e. the determination of activity releases in view of given design loads, in a detailed approach using cemented waste products under the action of mechanical stresses as an example.

CHARACTERIZATION OF CEMENT PRODUCTS

Today, cemented waste products are the most common form of conditioned wastes from the nuclear industry. The main reasons for cementing are the economy of this process of conditioning and the high level of safety thus attained. Examples of a few types of wastes which are usually fixed in a cement matrix today include:

- liquid wastes and sludges
- filter and evaporator concentrates
- combustion and pyrolysis residues
- incinerated solids
- building wastes and sand blast material.

The wastes are stirred into cement paste with which they form a firm homogeneous product after setting. Its compressive strength depends on kind and percentage of the foreign matter in the cement matrix and on the grade of cement chosen. In general, it is between 10 and 60 N/mm².

Mechanical stresses lead to a destruction of the waste product if the impact energy exceeds certain threshold levels. The waste product is then comminuted into a system of particles whose size distribution typically includes the following ranges:

(1) Range of very fine particles which can be dispersed as airborne particles (up to about 100 µm)

(2) Range of small particles which, as a result of the high rate of sedimentation, can only be dispersed at short distances up to about 20 m, depending on their initial impulse (about 100 to 1,000 µm)

(3) Range of large particles remaining at the place where the stress is applied (particles exceeding 1,000 µm).

Only the first range of particles is relevant to the accidental release of activity.

Cement products are frequently packed in receptacles for which no particular requirements for resistance against mechanical stresses have to be met. However, even such a receptacle will act as a barrier in the case of damage caused by an incident so that only a small percentage of the particles capable of dispersion are actually released. A quantification of this release-mitigating effect can only be carried out in special cases and is, in general, associated with a great deal of time-consuming effort.

However, since it is possible to conceive scenarios involving the release of the entire amount of dispersable particles - e.g. in the case of severe damage to the cover area of a drum - the following discussion will neglect the barrier effect of receptacles. Thus, the size distribution of the particles is the major information required for the determination of the activity release.

EXPERIMENTAL INVESTIGATIONS RELATING TO THE RELEASE OF ACTIVITY FROM CEMENTED PRODUCTS

At present, the determination of distribution functions in the case of mechanical stresses is generally carried out experimentally. Figure 1 illustrates the results of various experimental investigations as mass cumulative frequencies. The numbers refer to Table I which contains the essential information about the various curves.

TABLE I

Expalnation Concerning Fig. 1

#	Specific Energy J/g	Product Investigated	Ref.
1	2.6	Carbonated Ash Cement Product (Portland Cement)	2
2	7.5	High-Alumina, Neat	3
3	2.6	Carbonated Ash Cement Product (Blast Furnace Cement)	2
4	2.6	Filter Concentrate Cement Product	2
5	2.6	Ash Decontamination Concentrate Cement Product	2
6	2.6	Ash Cement Product	2
7	2.6	Filter Concentrate Cement Product	2
8	0.42	Resin Cement Product	4
9	0.2	Resin Cement Product	4

In experiments 1 through 7, samples are comminuted in a mortar by dropping weights. Sample materials and communition energy are the parameters.

Reference 4 reports on dropping tests carried out with cemented waste packages. Dropping height (5m - 43 m), percentage of foreign matter in the cement matrix (10% - 20%), size of package and type of packaging are the parameters.

Figure 1 shows that the distribution functions measured are considerably influenced by experimental boundary conditions such as type of waste and impact energy. This means that experimental investigations supply meaningful results only with respect to the specific boundary conditions in which the experiment was carried out. Thus curves 3 and 9 in Fig. 1 show the distribution functions for the 43 m and 20 m dropping tests of spherical resin products. However, without a suitable theoretical model it is impossible to derive the distribution functions for a 30 m drop from the former two functions.

The restricted significance is particularly disadvantageous in the planning phase of a facility during which frequent changes in the design of the facility, and thus the design loads, occur for a variety of reasons. In order to avoid carrying out new experimental investigations in the accompanying incident analysis whenever the plan is changed, it was necessary to determine accidental releases in a theoretical approach. For this purpose, a semi-empirical method was derived which permits a determination of distribution functions of particles as a function of the impact energy.

A THEORECTICAL METHOD FOR THE DETERMINATION OF THE ACTIVITY RELEASED FROM CEMENT PRODUCTS

To describe this method, the theoretical bases of comminution processes and distribution functions of systems of particles are dealt with first.

From a physical point of view, a comminution process is an energetic problem where energy is needed for the generation of new boundary surfaces. This is illustrated in Fig. 2 where the comminution of quartz according to Ref. 1 is used as an example and which shows the specific surface increase as a function of the specific comminution energy. The hatched areas are measured ranges, whereas the unbroken curve represents a calculated function. In Fig. 2, this energetic approach is confirmed by the fact that the newly generated comminution surface is proportionate to the impact energy over a large range of energy. This physical regularity is the basis of the theoretical method for the determination of the size distribution of particles in the case of a mechanical impact.

The further steps follow from the arithmetic of distribution functions.

As will be illustrated later on, particle spectra due to mechanical impacts can be sufficiently approximated by log-normal distributions in the radiologically relevant range. In Fig. 3, a typical log-normal distribution of a particle system is plotted on log-normal probability paper as size and area distributions. The following characteristic quantities of this distribution are still needed for further derivations:

count median diameter (d_g)	central value, i.e. 50% of the particles are below and 50% above this value
diameter of average area or mean surface diameter (d_a)	diameter at which the particle has the arithmetic mean of the surface
diameter of average mass or mean weight diameter (d_m)	diameter at which the particle has the arithmetic mean of the mass
mass median diameter (d'_m) (not plotted)	central value of mass distribution, i.e. 50% of the mass is below and 50% above this value

The geometric standard deviation σ_g an be derived directly from the representation of log-normal distributions.

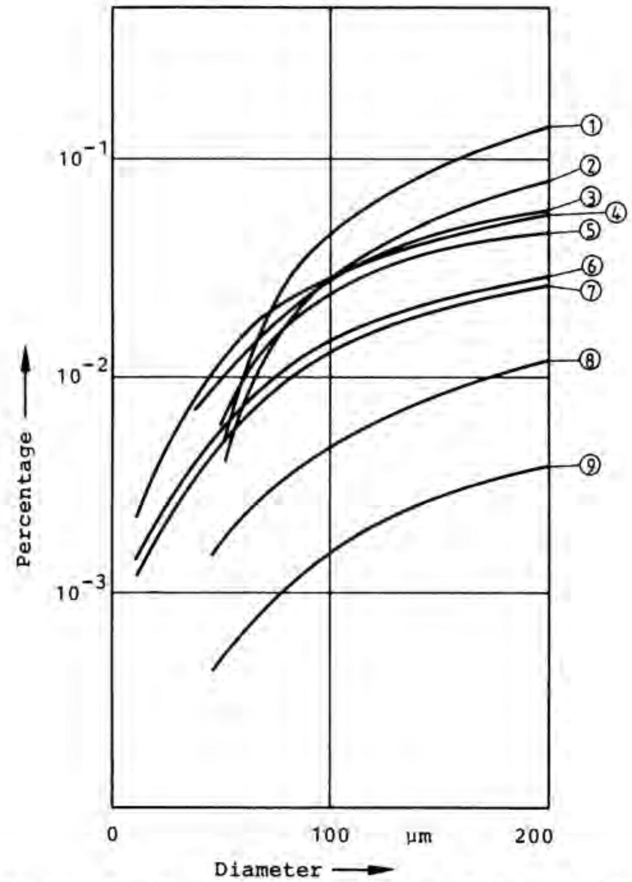

Fig. 1. Results of various experimental investigations comminution of concrete products.

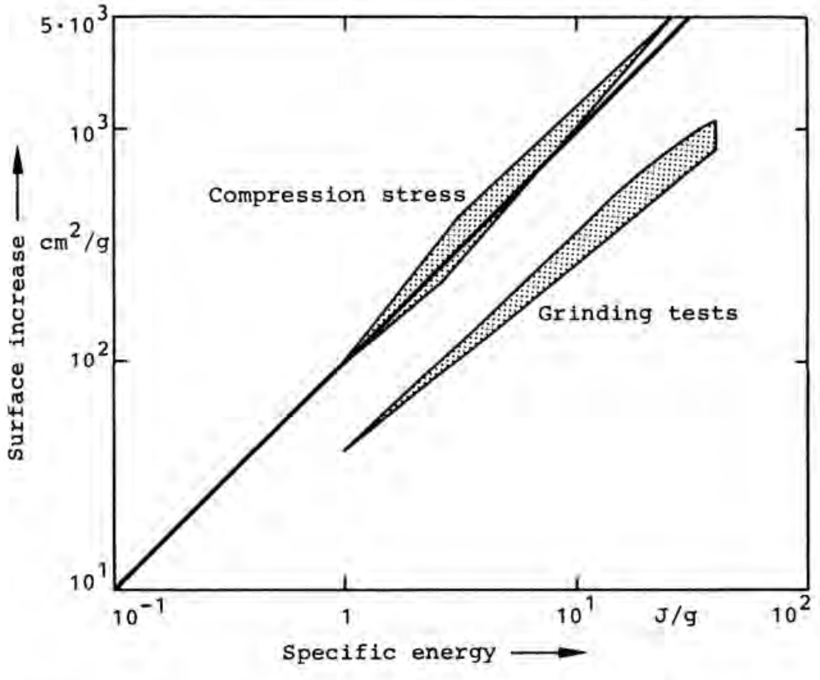

Fig. 2. Relationship between surface increase and impact energy[1].

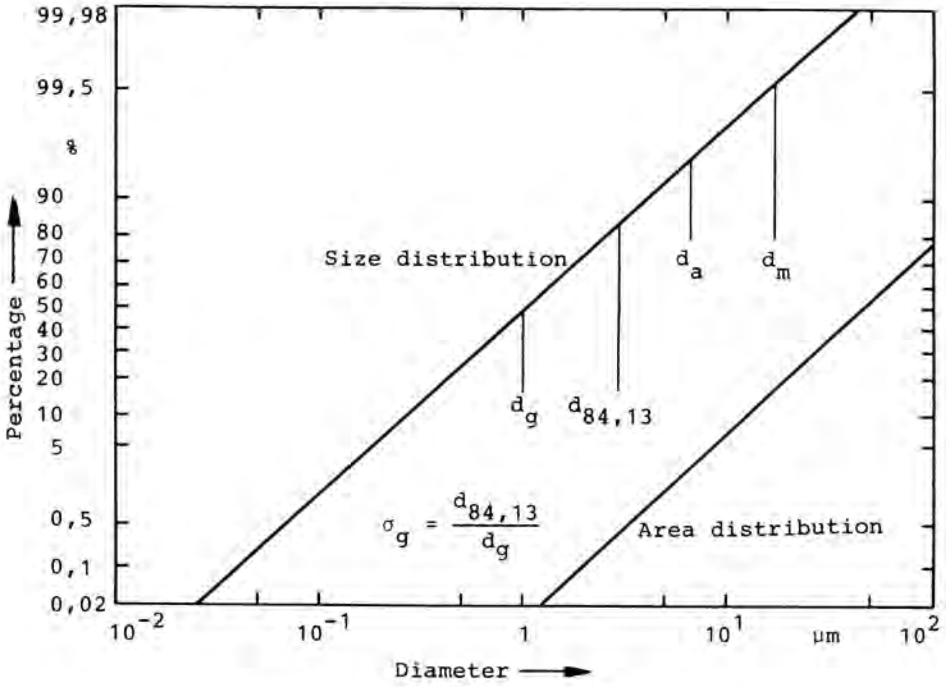

Fig. 3. Typical log-normal distributed size and area distribution plotted on log-normal probability paper.

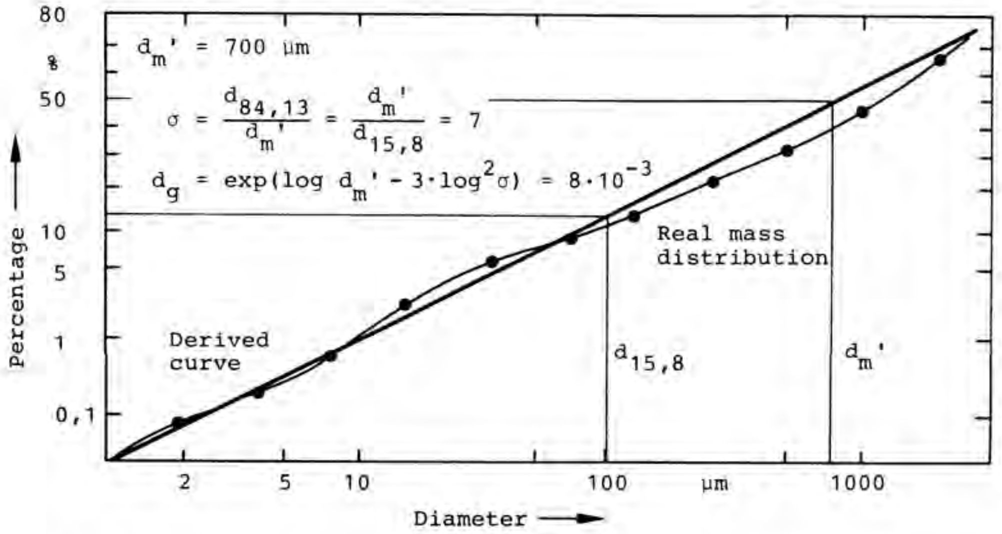

Fig. 4. Derivation of characteristic parameters of a log-normal distribution from an experimentally determined mass distribution.

$$\sigma_g = \frac{d_{84,1}}{d_g} = \frac{d_g}{d_{15,9}} \tag{1}$$

The following relationships exist between the quantities referred to above:

$$\log d_a = \log d_g + \log^2 \sigma_g \tag{2}$$

$$\log d_m = \log d_g + 1.5 \log^2 \sigma_g \tag{3}$$

$$\log d'_m = \log d_g + 3 \log^2 \sigma_g \tag{4}$$

If a collective of particles consists of n particles, its total mass is

$$m = \frac{n}{6} \pi \rho d_m^3 = \frac{n}{6} \pi \rho (\exp(\log d_g + 1{,}5 \log^2 \sigma_g))^3 \tag{5}$$

and the total area is

$$A = n \cdot \pi d_a^2 = n\pi (\exp(\log dg + \log^2 \sigma_g))^2 \tag{6}$$

Thus, the mass-related area is

$$A^* = \frac{A}{m} = \frac{6(\exp(\log^2 \sigma_g))^2 d_g{}^2}{\rho(\exp(1{,}5 \log^2 \sigma_g))^3 dg^3} \tag{7}$$

Following the presentation of these general theorectical bases, the approach for the determination of energy-dependent particle distributions is described using a practical example. Figure 4 shows a mass distribution determined experimentally [3]. For this real distribution, the log-normal distribution was derived. In a log-normal probability representation, this is the regression line. Thus, Fig. 4 confirms the previous statement that such distributions can be approximated sufficiently well by log-normal distributions.

From this log-normal distribution, the mass median diameter d'_m can be read directly as a 50% fractile. The geometric standard deviation σ_g results as the ratio between d'_m and the 15.8% fractile $d_{15.8}$ which can also be read directly. Using the two quantities, it is possible to determine d_g according to Eq. (4) and thus the specific area A* according to Eq. (7). A* is 290 cm². In relation to the specific impact energy of 10.3 J/g, the energy-dependent surface increase is obtained. In our example, a value of 28 cm²/J results.

A great number of experiments were evaluated this way. Over large energy ranges, the geometric standard deviation was found to be almost energy-independent. It is now possible to solve the problem referred to at the beginning, i.e. to convert a given distribution function of particles to other impact energies.

- The new specific surface to the collective of particles is derived from the energy-dependent surface increase and the new impact energy.
- The geometric mean d_g of the new distribution function is obtained by rearranging Eq. (7) and using the experimentally determined geometric standard deviation.

Using this method, the results plotted in Fig. 1 were evaluated and converted to a uniform energy of 0.05 J/g, cf. Fig. 5. Apart from the deviations of curves 1 and 6 or 7, there is a narrow range of distribution functions which is typical for cement products.

The values of energy-dependent surface increases are located between 5 and 35 cm²/J, those of the geometric standard deviation σ_g between 6 and 10.

The different courses of distribution functions 1 and 6 or 7 are a result of special properties of the material of these samples which are atypical for the behavior of cement products, e.g. a compressive strength that is too low.

The uniform behavior of the distribution function is both surprising and a good verification of the theoretical approach presented here, insofar as the experiments were carried out with most diverse boundary conditions, as mentioned before.

If one is faced with the task of theoretically determining, in advance, activity percentages released from an unknown cement product with given design loads, the following values will provide a distribution function covering the radiologically relevant particle spectrum up to 100 µm:

energy-dependent surface increase - 40 cm²/J
geometric standard deviation - 7

If these parameters are used to calculate, for example, the particle distribution of a cemented waste package following a 60 m drop, it results that about 2 x 10^{-4} per cent weight are smaller than 125 µm. For this case, a scale experiment [5] determined a value of 1.2 x 10^{-4} by sieve analysis. This means that the theoretical approach presented supplies sufficiently realistic results and is thus a suitable tool for the determination of accidental activity releases.

SUMMARY

In order to be able to determine the accidental release of radioactivity from waste packages, it is necessary to know the distribution of the particles of the product which results from mechanical stresse. Presently, a great number of individual experiments relating to the communition behavior of cemented products are available. A transfer of these data to other boundary conditions such as they may occur in a final repository as a result of technical characteristics, has so far not been possible.

For the solution of this problem the present paper discusses a method permitting the theoretical determination of activity releases from cement products on the basis of given stresses within the scope of incident analyses.

For verification purposes, this method was first used for the conversion of various experimentally determined particle spectra to uniform boundary conditions. The results indicate that cement products show a typical comminution behavior within narrow bounds. The converted distribution functions permit a derivation of parameters which can also be used to calculate the release from cement products for which experimental investigations have not yet been carried out.

REFERENCES

1. Rumpf, H.
Zerkleinern
in: Ullmanns Encyklopädie der technischen Chemie, Band 2, Ausgabe 1972
Verlag Chemie, Weinheim (1972).

2. NUKEM GmbH
Neue Ergebnisse zur Freisetzung aus radioaktiven Abfällen bei mechanischer und thermischer Belastung
Anhang zu FUE 84010, Hanau, 1984

3. Wallace, R.M., et al.
An Impact Test for Solid Waste Forms
E.I. Du Pont de Nemours and Company
Savannah River Laboratory
Aiken, South Carolina
March 1976

4. Rämö, E., et al.
Damage of Reactor Waste Packages in Transport Accidents
NKA, AO (79) 17, 1979

5. Vejmelka, P., et al.
Activity Release from Waste Packages Containing LLW-Cemented Waste Forms Under Mechanical and Thermal Stresses
CEC-Seminar: Testing, Evaluation and Shallow Land Burial of Low and Medium Radioactive Waste Forms
Geel, Belgium, 28-29 September, 1983.

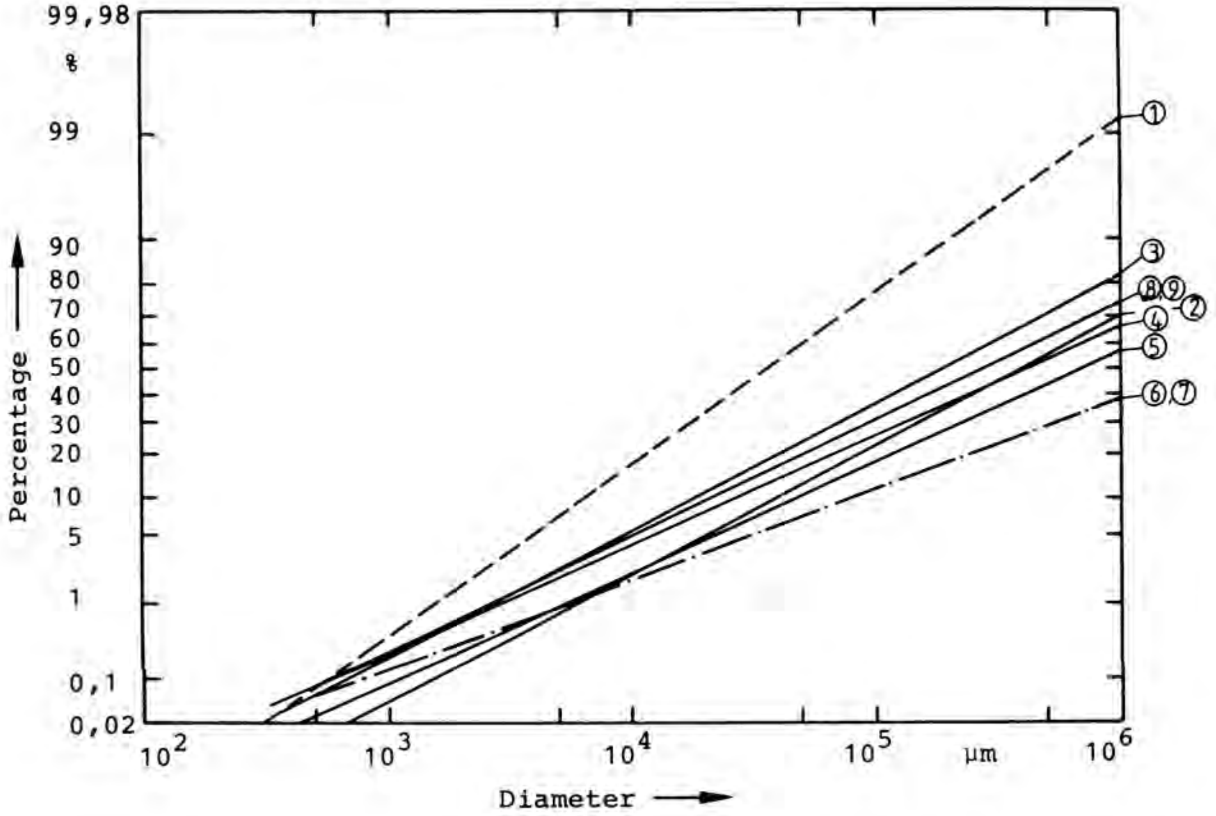

Fig. 5. Mass distribution of various experimental investigations after conversion to a standard impact energy.