Accidental Release from Radioactive Waste Packages

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ABSTRACT

In order to protect the environment against pollution by hazardous materials in the course of a shipping accident, such materials have to be conditioned in waste packages. For this purpose, the hazardous materials are either packed into a high-quality container or immobilized within a matrix. The calculation of the accidental release of hazardous materials is an essential factor in the assessment of transportation hazards. The release processes depend on the forces acting on the cargo and on the physical properties of the packages and of the product itself. The purpose of this paper is to demonstrate the influence of the properties of the product on the release of hazardous materials. This aspect is discussed with respect to the accidental release from hazardous wastes that are immobilized (for example, cemented wastes) and from those wastes that are not immobilized.

The transportation of radioactive wastes belongs to the category of the transportation of hazardous materials involving certain risks. Risk and safety analyses are carried out for the quantification, limitation, and minimization of these risks.

An essential question to be answered within the scope of these safety analyses is that of the amount of pollutants that may be released to the environment in an accident. During the transportation of radioactive waste packages, this is a function of the severity and kind of stresses occurring during an accident as well as of the packaging of the waste and the waste form itself. With respect to the hazard inherent in the waste inventory, which becomes acute when radioactive materials are released, the immobilization of the inventory in a solid matrix (waste form) and the packaging constitute the decisive barriers for the limitation of the hazard. The principle of this correlation is shown in Figure 1.

The barrier effect of the waste form itself and of the solid matrix is discussed with a view to the limitation of the release of pollutants in the event of an accident. It is also demonstrated that the results obtained can be applied to the transportation of other hazardous materials, provided that the solid matrices investigated are comparable with these materials.

CLASSIFICATION OF RADIOACTIVE WASTES IN TRANSPORTATION

In 1985 the International Atomic Energy Agency (IAEA) issued new regulations for the transportation of radioactive wastes (1). Similar to the regulations already in force, the new regulations also provide for a classification of the radioactive materials in accordance with the kind and amount of their activity and their radiological properties and, for an allocation to certain classes of transportation, with the barrier effects of their packaging. Figure 2 is a simplified representation of this classification.

Radioactive materials with high levels of activity and radiotoxicity are transported in Type B packages; those with medium levels of activity are transported in Type A packages. Industrial packages are used for the transportation of radioactive materials with low levels of activity and radiotoxicity, for example, those that are uniformly distributed in a solid matrix [low-specific-activity (LSA) and SCO materials].

Depending on the different hazard potentials of the radioactive materials, the packages have to meet different requirements as far as their barrier effects are concerned. In the case of Type B packages, the barrier function in transportation accidents is assumed by the packaging alone. This packaging has to be designed and manufactured in such a way that even the most serious accidents will not cause any unacceptable alteration of the barrier effect. Type B packages need not meet any specific requirements with respect to waste form and limitation of amount of activity.

In Type A packages, the barrier function in the case of transportation accidents is assumed by the packaging and in part by the waste form. The amount of activity is limited.

In industrial packages, materials transported are limited to those with low activity. The requirements for packaging and waste form pertain to handling and operation and not to accidents. Accordingly, no barrier function is allocated to either the packaging or the waste form.

The overwhelming majority of radioactive wastes that result from waste management at nuclear power plants are in the category of low activity. As far as these wastes are concerned, the radioactive

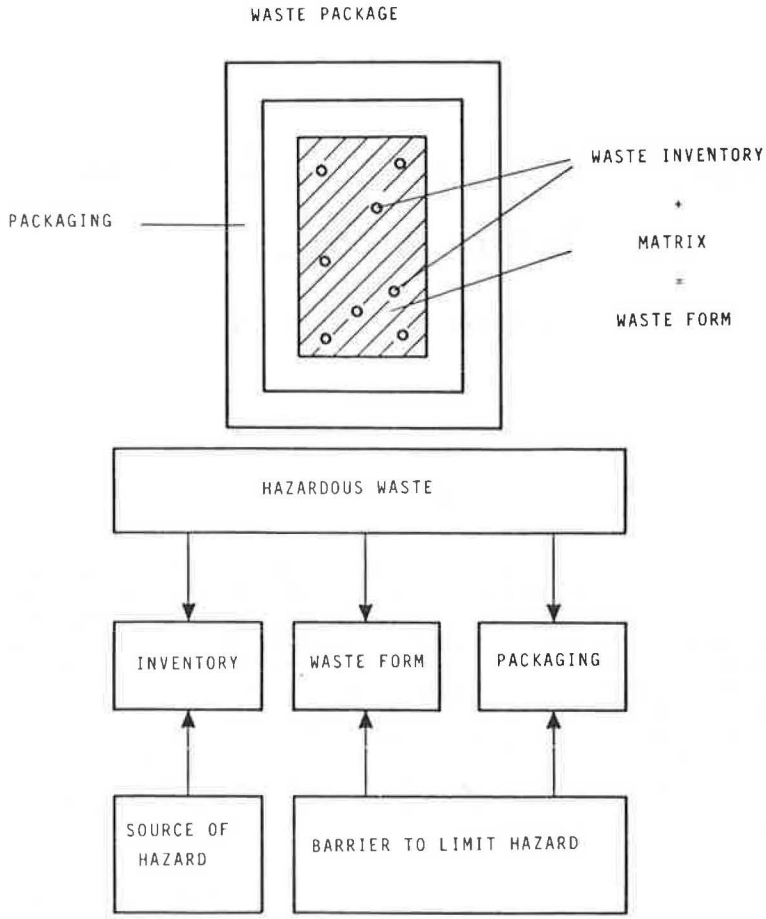

FIGURE 1 Characterization of hazardous waste.

materials are either immobilized in a solid matrix or attached as contaminations to solids. In the sense of the IAEA regulations, they have to be classified as LSA or SCO materials. Accordingly, more than 90 percent of the wastes produced at nuclear power plants can be transported in industrial packages (2).

Although the transportation regulations provide that packaging and waste form have to fulfill only a moderate function with respect to the mitigation of release, the waste form has a considerable mitigating effect in practice. As a result of its physical and chemical solid properties, it is capable of limiting the release of pollutants when it is sub-

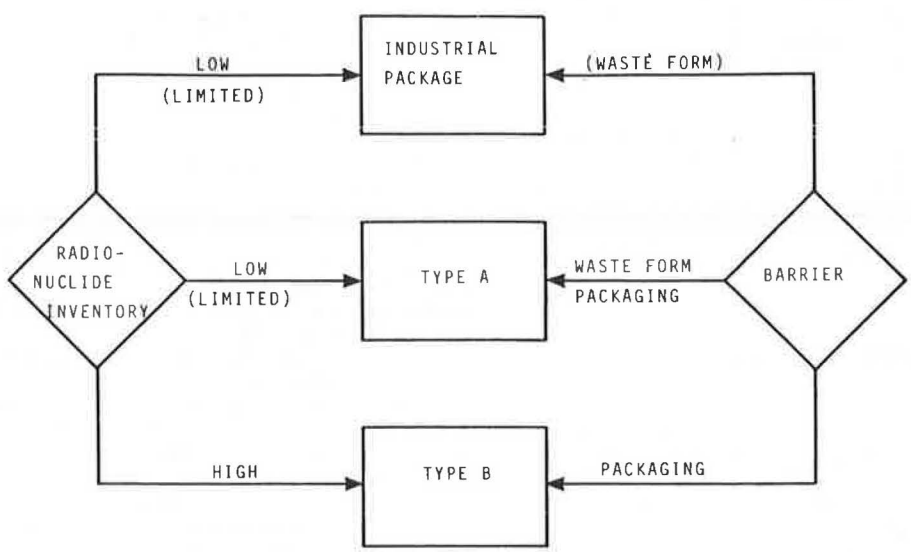

FIGURE 2 Classification of radioactive waste packages according to IAEA regulations.

jected to a mechanical stress. This barrier effect has to be considered when the release of pollutants is determined by risk analyses. Thus, the following is a discussion of how this barrier effect can be quantified.

QUANTIFICATION OF STRESSES ACTING ON WASTE FORM IN A TRANSPORTATION ACCIDENT

The majority of the stresses that may occur during a transportation accident and lead to release of wastes from a waste package are mechanical or thermal or both. In practice, mechanical stresses have to be anticipated in all conceivable accident scenarios, for example, in a collision of vehicles in road transportation, in rear-end collisions, or in a derailment of a railroad car. Because of fuel fires, thermal stresses may occur in particular in accidents during road transportation.

The following discussion is restricted to purely mechanical stresses, because comparable investigations concerning thermal stresses have not yet been completed (3).

The mechanical stresses encountered in accident situations can be traced mainly to three fundamental types:

1. The impact of a waste package on a flat obstacle,
2. The impact of a waste package on a pointed obstacle, and
3. The crushing of a waste package.

Figure 3 is a schematic representation of these three types of stresses.

The differentiation between stress types 1 and 2 is important if the packaging assumes an essential barrier function. In such a case, the dropping of a waste package may result in its penetration by the pointed obstacle, whereas the packaging will withstand the first type of stress, although the height of fall is the same for both types. The differentiation between types 1 and 2 is no longer of any significance if the stresses are so great in relation to the packaging barrier that a dramatic failure of the packaging has to be anticipated in either case.

With respect to a negligible barrier effect of the packaging, all three types of stresses are reduced to one quantity, namely, the specific energy input into the waste form. This means that the mechanical stress occurring in an accident may be modeled by the specific energy input into the waste form in order to determine the release of pollutants from immobilized low-activity wastes, that is, materials transported in industrial packages as shown in Figure 2.

Figure 4 provides a survey of the possible range of the specific energy input in different accident scenarios. The figures given apply, for example, to a 200-L hooped drum containing cemented waste materials and weighing 500 kg on the conservative assumption that the energy input into the waste package during the various accident situations will occur without any loss. For example, in the case of a rear-end collision in which a 20-ton truck impacts the rear of the transportation vehicle at a speed of 50 km/hr, the waste package is subjected to the maximum crush stress that may occur in such an accident. Figure 4 shows that very high-energy inputs may occur, particularly in accident situations where crush stresses are caused by the mass of a multiple of the weight of the waste package. It should be noted that these calculated stresses do not take into account the shock absorption of the vehicles, and that real stresses in real accidents would be less than the calculated values.

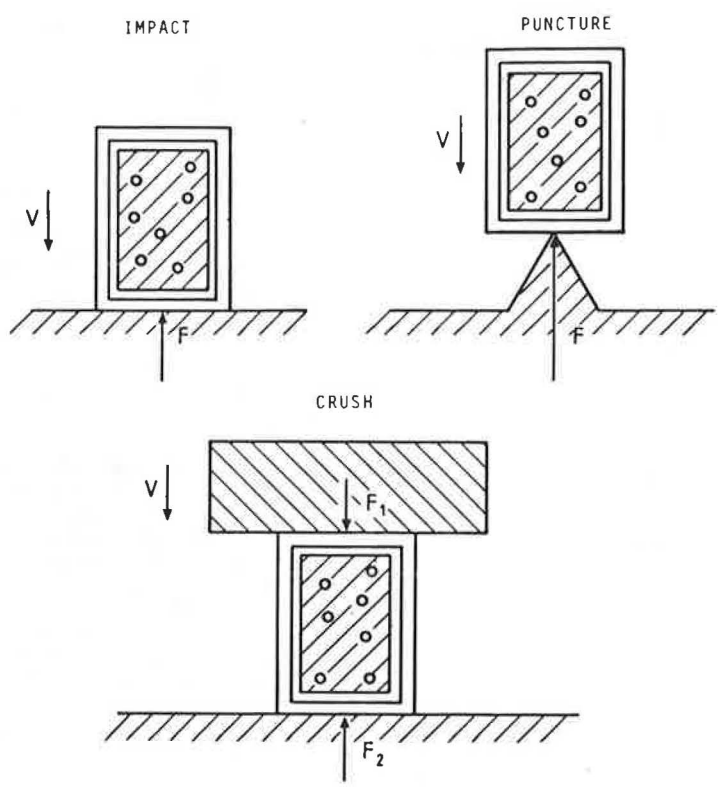

FIGURE 3 Mechanical stresses on waste packages in transportation accidents.

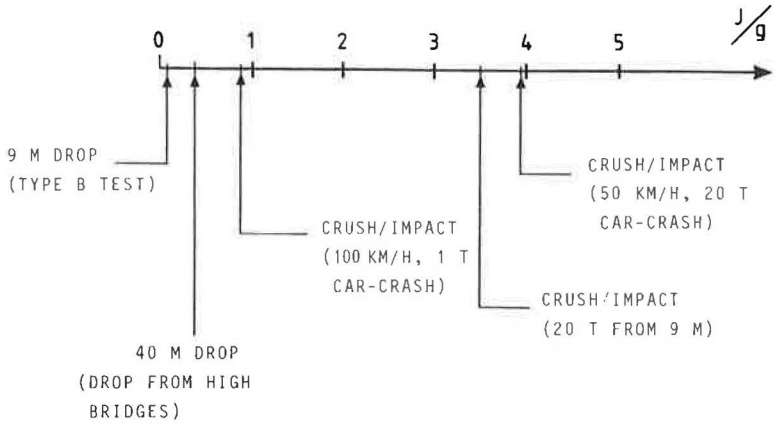

FIGURE 4 Energy input into 500-kg waste package caused by mechanical stress in different accidents.

QUANTIFICATION OF RELEASABLE AMOUNTS OF POLLUTANTS

The determination of the release of radioactivity is described in detail by using cemented waste products as an example. Today cemented waste forms are the most common type of conditioned waste from the nuclear industry. The waste is stirred into cement paste with which it forms a firm homogeneous product after setting.

Mechanical stresses lead to a destruction of the waste form if the impact energy exceeds certain threshold levels. The waste form is then comminuted into a system of particles whose size distribution typically includes the following ranges (Figure 5):

1. Very fine particles that can be dispersed as airborne particles (up to about 100 μm);
2. Fine particles that, as a result of the high rate of sedimentation, can only be dispersed for short distances, depending on their initial impulse (about 100 to 1000 μm); and
3. Large particles remaining at the place where the stress is applied (particles > 1000 μm).

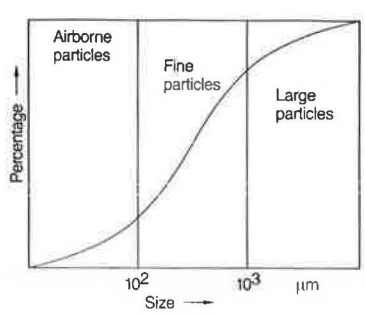

FIGURE 5 Cumulative mass distribution.

Only the first category of particles is concerned in the accidental release of pollutants. Thus, the size distribution of the particles as a function of the impact energy is the major information required for the quantification of the amounts of pollutants that can be released. From a physical point of view, this comminution process is an energy problem in which energy is needed for the generation of new boundary surfaces. Comminution experiments show that the newly generated surface is proportional to the impact energy over a large range (4). 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.

As will be illustrated later, particle spectra due to mechanical impacts can be sufficiently approximated by log-normal distributions in the range relevant to release. In Figure 6, a typical log-normal distribution of a particle system is plotted on log-normal probability paper as size and area

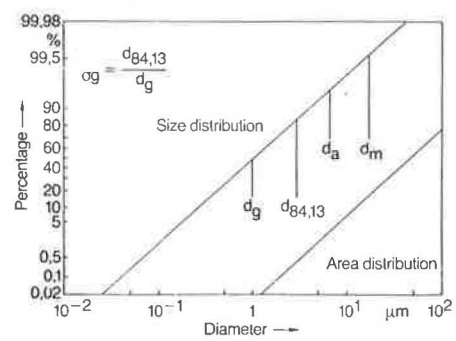

FIGURE 6 Log-normal distributed size and area distribution.

distributions. The following characteristic quantities of this distribution are still needed for further derivations:

1. Count median diameter (d_g): the central value; that is, 50 percent of the particles are below and 50 percent above this value;
2. Diameter of average area or mean surface diameter (d_a): the diameter at which the particle has the arithmetic mean of the surface;
3. Diameter of average mass or mean weight diameter (d_m): the diameter at which the particle has the arithmetic mean of the mass; and
4. Mass median diameter (d'_m) (not plotted): the central value of mass distribution; that is, 50 percent of the mass is below and 50 percent above this value.

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

$$\sigma_g = d_{84.13}/d_g = d_g/d_{15.87} \quad (1)$$

The following relationships exist between the quantities just listed:

$$\log d_a = \log d_g + \log^2 \sigma_g \quad (2)$$

$$\log d_m = \log d_g + 1.5 \log^2 \sigma_g \quad (3)$$

$$\log d'_m = \log d_g + 3 \log^2 \sigma_g \quad (4)$$

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

$$m = (n/6)\pi\sigma d_m^3 = (n/6)\pi\sigma\,[\exp(\log d_g + 1.5 \log^2 \sigma_g)]^3 \quad (5)$$

The total area is

$$A = n\pi d_a^2 = n\pi\,[\exp(\log d_g + \log^2 \sigma_g)]^2 \quad (6)$$

Thus, the mass-related area is

$$A^* = A/m = \{6\,[\exp(\log^2 \sigma_g)]^2 \times [\exp(\log d_g)]^2\} / \{\sigma\,[\exp(1.5 \log^2 \sigma_g)]^3 \times [\exp(\log d_g)]^3\} \quad (7)$$

Following the presentation of these general theoretical bases, the approach for the determination of energy-dependent particle distributions will be described by using a practical example. Figure 7 shows a mass distribution determined experimentally (5). For this real distribution, the log-normal distribution was derived. In a log-normal probability representation, this is the regression line. Thus, Figure 7 confirms the previous statement that such distributions can be approximated sufficiently well by log-normal distributions.

From this log-normal distribution, the mass mean diameter d_m' can be read directly as a 50 percent fractile. The geometric standard deviation σ_g results as the ratio between d_m' and the 15.8 percent fractile $d_{15.8}$, which can also be read directly. By using the two quantities, it is possible to determine d_g according to Equation 4 and thus the specific area A* according to Equation 7. Accordingly, A* equals 290 cm^2. In relation to the specific impact energy of 10.3 J/gm, the energy-dependent surface increase is obtained. In this example, a value of 28 cm^2/J results.

A great number of experiments were evaluated in this way (6). Over large energy ranges, the geometric standard deviation was found to be almost energy independent. It is now possible to convert a given distribution function determined at a certain impact energy to other impact energies:

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

For the cemented wastes discussed here, the values of the energy-dependent surface increase are between 5 and 32 cm^2/J and those of the geometric standard deviation between 6 and 10 cm^2/J.

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

1. Energy-dependent surface increase, 40 cm^2/J;
2. Geometric standard deviation, 7 cm^2/J.

If these parameters are used to calculate, for example, the particle distribution of a cemented-waste package following a 60-m drop, the result is that about 0.2 percent by weight is smaller than 125 µm. This value has been confirmed by scale experiments (7).

If a homogeneous distribution of the activity in the waste form is assumed, the share of particles having a diameter < 100 µm corresponds to the share of activity released. This means that the immobilization of the waste constitutes a barrier retaining more than 99 percent of the activity in the case of a 60-m drop.

The mitigating effect on the release of activity of immobilizing the waste becomes particularly obvious if a comparison is made with the release behavior of nonimmobilized material. The latter often has a significant share of dispersible particles, which is only revealed as a result of a mechanical impact. Figure 8 shows particle spectra of powdered resins and combustion residues (8,9) as examples of unfixed wastes containing a high dispersible share.

It it is assumed, as in the case of the cement

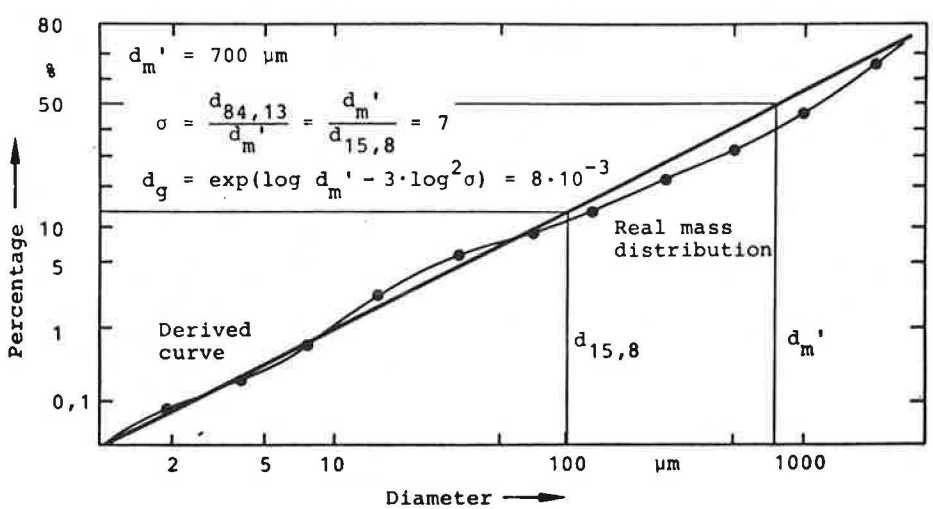

FIGURE 7 Derivation of characteristic parameters of a log-normal distribution from an experimentally determined mass distribution.

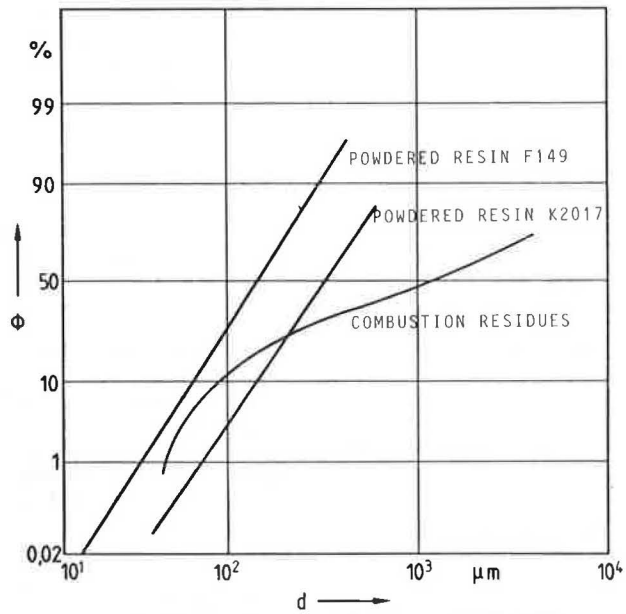

FIGURE 8 Cumulative mass distribution curves of various powdered wastes.

products, that the share up to about 100 μm is released, the quantities of pollutants that may be released amount to up to 30 percent. This means that even highly dispersible systems possess a barrier effect preventing a 100 percent release. By a fixation of the waste, the barrier effect of the waste form is improved to such an extent that the share that can be released is reduced by a factor of about 100.

SUMMARY

In the transportation of radioactive wastes, the protection of the environment against such materials plays an important role. The determination of the extent of the hazard potential, particularly in accidents, is carried out by means of safety analyses. These analyses also indicate which measures may be taken to mitigate the risk.

The calculation of the release of pollutants as a function of an energy impact was demonstrated by using as an example radioactive powders and ashes fixed in a cement matrix. It was demonstrated that in a serious accident the release of pollutants is reduced by a factor of about 100 for waste fixed in a cement matrix compared with waste unfixed in a packaging that is damaged in the course of the accident.

In this respect, the analyses presented may also serve to initiate the technical realization of the requirements to be met for the confinement of pollutants. However, some further analysis will have to be used to find out to what extent detailed requirements for the quality of packaging may be replaced by fixation of the pollutants in a solid matrix. The discussion here centers on the possibility of quantifying the barrier effect during an accident of such fixation.

The validity of the results presented is not limited to a release of radioactive materials. The method presented may be applied whenever pollutants are uniformly distributed and fixed in a solid matrix , or are themselves solids, and may be released as attachments to the resulting dust particles when the matrix is destroyed mechanically.

Above all, metallurgical slags and dusts with heavy metals attached as well as catalysts, distillation products, and other wastes of chemical conversion and synthesis processes should be mentioned here.

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