DETERMINATION OF PROBABILISTIC SOURCE TERMS FOR WASTE DISPOSAL FACILTIES

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ABSTRACT

Probabilistic Safety Analysis (PSA) has proved to be a useful tool for demonstrating the high safety level of Nuclear Power Plants (NPP). However, the methods developed for NPP could not be applied for disposal facilities. With respect to the operational phase of a final repository for radioactive wastes a special approach has been developed taking release behavior of radioactive waste into account. The total spectrum of Low Activity Waste (LAW) and Intermediate Level Waste (ILLW) is grouped into waste streams of similar characteristics. Distributions of release fractions are determined for these distinct waste streams as a function of the incident loads and the waste properties.

These distributions are especially suitable for the application in Monte-Carlo-Simulations. In addition to “rolling dice” regarding the selection of waste, activity inventory and incident scenarios, the release fraction is likewise determined in a realistic way by “rolling dice”. Thus a realistic dose distribution can be determined, which is essential for the adequate safety design of a facility. After determining the probabilistic source term distributions, the PSA can easily be carried out with minor expenditures in terms of costs and time.

OBJECTIVE

The derivation of waste acceptance criteria is usually supported by a deterministic safety analysis. Within the framework of these analyses deterministic source terms are determined. The deterministic source term determination must cover all cases with respect to load assumptions and material composition. Thus conservative release fractions - and in turn conservative acceptance criteria - are assessed.

Waste acceptance criteria that are more realistic result from Probabilistic Safety Analysis (PSA). In order to be able to carry out a PSA, a new approach for a determination of probabilistic source terms had to be developed. The approach taken accounts for the entire load spectrum and the variety of materials and results in a spectrum of release fractions as a function of various influence parameters.

In this presentation we will describe the determination of probabilistic source terms on the basis of the example of a rectangular steel container with cemented waste in a fire accident (The complete approach for the determination of probabilistic source terms considers of course the entire spectrum of packages). A short overview on the application in a PSA is also given. The possible influence on waste acceptance criteria will be discussed.

APPROACH TAKEN IN DETERMING THE PROBABILISTIC SOURCE TERM

In a deterministic determination of source terms maximum release fractions are calculated for specific design loads (e. g. in the case of fire accidents the maximum fire load). The maximum source term results from conservative parameters within a spectrum of parameter relevant to the release spectrum, e. g. the chemical compound with the highest vapor pressure. Consequently the deterministic source term is one distinct value.

In a probabilistic determination of source terms on the other hand paired values consisting of release fractions and their respective probabilities or a continuous probabilistic distribution of the release fraction are calculated. In general this requires a large number of calculations in which the variability in real waste characteristics and the probabilistic distributions of parameters relevant to the release are taken into consideration. Typical parameters influencing for the probabilistic determination of source terms relate to the properties of the waste product, of the waste package, of the incident scenarios and other related factors.

The probabilistic distribution of certain properties of the waste package, such as the percentage of burnable material, can be determined by a statistical analysis of the entire spectrum of waste packages. The probabilistic distribution of the load spectrum results from the respective incident analysis.

In some cases the distribution functions may be quite simple, e.g. a constant or linear function. However, given pre-determined discrete values and scenarios the related probabilistic distributions may provide lead to irregular, complex equations, which are difficult to deal with mathematically. With the help of curve fitting procedures, these complex distributions can be converted to mathematically more convenient functions. The Gaussian error function in accordance with equation (1) is one such function which covers most of the spectra to be considered.

$$w = \frac{const \cdot 2}{\sqrt{2}} \cdot \int_{-\infty}^{\frac{1}{\sqrt{2}} \bullet \frac{\ln(\frac{x}{\hat{x}})}{s}} \exp(-t^2)\,dt \qquad (1)$$

The parameters const, $\hat{x}$ and sigma are used to fit the curve to the real values. Using this function, any source term distribution can then be defined by only three parameters. Figure 1 shows an example of a complex distribution of release fractions and its Gaussian error function curve fit. It is clearly seen that the over- and underestimations of the fitted curve tend to compensate each other and that this curve is, thus, a satisfactory average distribution. The distribution accounts both for the probability of the scenarios and the probability of waste properties and also takes into account the release behavior of the waste.

In the next section this approach is described in detail using a typical waste composition and an exemplary accident involving a fire.

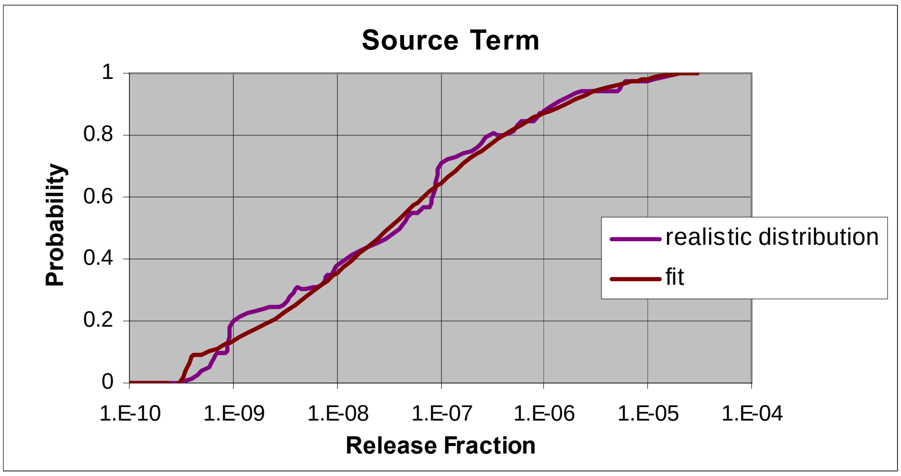

Fig. 1: Fit of a real release distribution by use of the Gaussian error function

PROBABILISTIC PARAMETER FOR DETERMINING THE SOURCE TERM

In earlier studies on the release behavior of radioactive wastes under thermal stress many release models have been developed which are all suitable for determining the source term for any combination of loads, material and package compositions. At the same time, incident analyses have lead to a wide spectrum of fire incidents with their respective frequencies and range of loads. The respective source terms are then calculated on the basis of the probability for a large number of distinct incidents. These source terms, then, lead to the desired probabilistic distribution.

Release Mechanisms Under Thermal Stress

There are four relevant release mechanisms resulting from thermal stress:

- release due to pyrolysis of the waste product,
- release due to combustion of the waste product,
- release due to vaporization of water in the waste product,
- release due to sublimation or vaporization of radioactive substances.

Pyrolysis

Pyrolysis is understood as the thermal decomposition of a material. In waste packages, this process takes place if the temperature of the product exceeds a certain threshold, e.g. approximately 300 °C, with a limited admission of air and, provided, the waste product contains thermally instable constituents. The gaseous decomposition products - the so-called pyrolysis gases - escape from the waste package and can burn outside of the package if ignited. A certain percentage of activity is released with the pyrolysis gases.

Experimental investigations have shown that the fraction of activity released by pyrolysis is approximately $5\ 10^{-3}$ with respect to cesium (1).

The fraction of released activity is smaller for less volatile nuclides.

Combustion

Combustible wastes will burn if a sufficient amount of air is available. A number of experimental investigations have been carried out to determine fractions of activity release due to combustion (2), (3), (4), (5), (6), (7). In these experiments, the release fractions determined were between 10^{-2} and 0.4 with the wide span of these results being due to different experimental boundary conditions.

The resuspension of the residues of combustion may also have to be added to these release fractions.

Vaporization of water

The vaporization of water also transports radioactive substances into the gaseous phase due to co-vaporization. This release mechanism is particularly relevant for cemented wastes and concentrates as these wastes contain great amounts of vaporizable water in their pore volume, water of crystallization, or the like.

The investigations of the evaporation of liquids (8) and (9) have shown that related release fractions lie in the order of 10^{-5} to 10^{-3}. The investigations of cemented wastes in 200-l-drums (1) and (10) have confirmed the results of the evaporation experiments.

Sublimation or vaporization of radioactive substances

In addition to the release of radioactive substances by means of co-vaporization, radioactive substances are also subject to direct vaporization or sublimation. In general, this is the dominant release pathway of volatile substances such as iodine or tritium. However, it also can be of importance to other elements, in particular cesium. The percentage of activity released depends on the duration and the temperature level of sublimation. The amount of activity released can be estimated theoretically on the basis of Lewis' law and the thermodynamic data of the substances involved.

Relevance of Release Mechanisms for Various Wastes

In the case of cemented wastes, a combination of the release mechanisms of pyrolysis (in hot surface layers) and co-vaporization must be considered. To quantify the individual contributions it is assumed that co-vaporization is the main process in those parts of the volume where the temperature is higher than 100 °C, whereas pyrolysis takes place in those parts of the volume where the temperature is higher than 300 °C.

As far as scrap is concerned, the only relevant process for the release of radionuclides is their direct sublimation/vaporization.

Compacted wastes may consist of pure scrap or scrap mixed with other media such as textiles and paper. In the case of a strongly metallic composition (scrap), the dominant release process is sublimation/vaporization of the radionuclides, in all other cases it is pyrolysis.

Apart from the above-mentioned mechanisms for the release of radionuclides, the release of easily volatilized compounds such as organic iodine is determined by considering sublimation in all kinds of waste flows.

With respect to cemented wastes, scrap and compacted wastes, usually, no combustion related release fractions need to be considered. This is because the integrity of packaging is generally preserved to such an extent that only pyrolysis takes place.

Load Assumptions

The released activity mainly depends on the load assumptions for thermal stress. In this context, thermal stress is always assumed to be due to the impact of a fire. For example, the load assumptions determine the volume percentages at temperatures of more than 300 °C or more than 100 °C, and/or the waste temperatures relevant to sublimation.

Usually, the load assumptions are stated as a temperature-time function for the fire. In order to calculate waste heat-up in the package, the heat transfer coefficients for both convection and radiation have to be quantified. Considering a realistic fire load, other factors have to be taken into account, e. g. to what extent the package is exposed to the fire, or whether the package is shielded by other objects or whether there are other heat sinks.

The kinetics of the fire is influenced by a number of parameters which include, among others:

- distribution and composition of the fire loads,
- radiation properties of confining walls,
- cooling of the fire by air draft,
- reduction of the combustion speed by a limited supply of air,
- unsteady temperature field in a flame.

It is impossible to consider all these parameters in the theoretical description of the temperature-time function.

Therefore, mean model curves are used in the realistical description of the effects of a fire on a waste package even though peak values of the expected realistic temperature-time history are not reached. A well-known example of such a model curve is the load assumption of 800 °C for a period of 30 minutes that is contained in the IAEA transport regulations. Another curve considering 800 °C for a period of 60 minutes is cited in (11) with respect to the accident covering all eventualities in the final disposal of radioactive wastes.

The latter model curve results from experimental investigations in technical scale. The long time period is mainly caused by the large amount of petrol. However, after filling up, the petrol volume decreases continuously. This leads to a fire duration between 20 and 60 minutes. In our example a linear time distribution between 20 and 60 minutes is assumed. Analogously, a probabilistic distribution of the temperature level is assumed from values lying between 600 °C and 800 °C. Both assumptions are in accordance with comparable realistic fire scenarios.

SOURCE TERM DETERMINATION

The approach for determining the source term is shown using a rectangular waste package with cemented waste. The source term determination is performed in several steps:

1. A distinct scenario is chosen.
2. Temperature fields are calculated for the distinct scenario.
3. Volumes with temperatures > 300 °C and >100 °C are determined from the temperature field calculation.
4. The release fraction for the chosen scenario is calculated by the use of the given release models.

The selection of the scenario is carried out by “rolling dice”: Two random numbers, r_1 and r_2, with values between 0 and 1 are generated by the computer. The resulting scenario is:

$$\textbf{Temperature [°C]} = 600 + r_1 * 200 \qquad (2)$$

$$\textbf{Time [min]} = 20 + r_2 * 40 \qquad (3)$$

With these parameters a temperature calculation is carried out. Figure 2 shows 12 selected temperature functions.

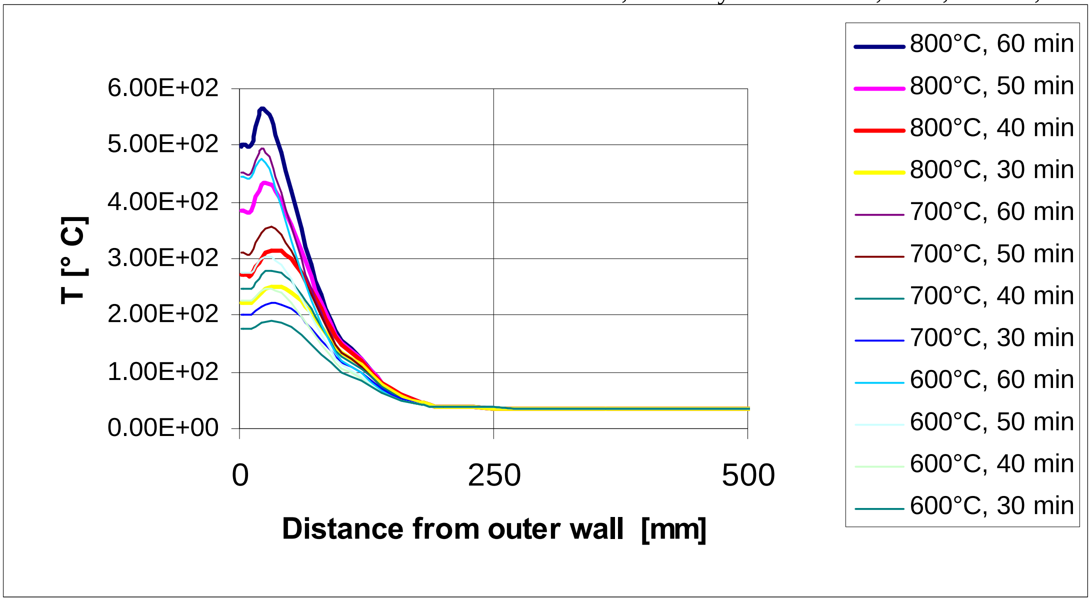

Figure 2: Temperatures as function of the heat penetration depth

With increasing fire times and fire temperatures the temperatures in the container increases. However, the penetration depth of the 100 °C-isotherm is hardly influenced by these parameters. Even at a fire temperature of 800 °C, the temperature in the waste product exceeds 300 °C only for fire times longer than 40 minutes. At a fire temperature of 600 °C, more than 50 minutes is needed.

In order to determine the 300 °C- and the 100 °C-isotherms, many temperature field calculations were carried out for the above mentioned time and temperature distributions

A summary of the temperature calculations is presented in Table 1. In addition to the calculated penetration depths for the temperature limits of 100 °C and 300 °C, the fractions of the volumes δV/V are given for which the temperature limits are exceeded. The last column shows the release fraction resulting from heating of the waste package which was calculated using equation (4).

$$\mathbf{F = dV/V(300\ °C) \cdot 5 \cdot 10^{-3} + dV/V(100\ °C) \cdot 5 \cdot 10^{-4}} \qquad (4)$$

The constants 5 10^{-3} and 5 10^{-4} are experimentally derived release fractions for pyrolysis and co-vaporization release mechanisms.

Table 1: Release fractions for a steel container with cemented waste in dependence of fire temperature and fire time

max. temperature	**time**	**penetration depth [mm]**		**dV/V**	**dV/V**	**release fraction**
°C	**[min]**	**300°C**	**100°C**	**300°C**	**100°C**	
800	60	68	132	0.11	0.098	5.00E-04
	50	62	131	0.10	0.11	4.64E-04
	40	51	130	0.085	0.12	3.98E-04
	30	0	128	0	0.20	8.36E-05
700	60	61	123	0.10	0.095	4.53E-04
	50	53	122	0.088	0.11	4.05E-04

max. temperature °C	**time [min]**	**penetration depth [mm]**		**dV/V 300°C**	**dV/V 100°C**	**release fraction**
		300°C	**100°C**			
	40	0	120	0	0.19	7.88E-05
	30	0	119	0	0.19	7.82E-05
600	60	56	115	0.093	0.091	4.19E-04
	50	32	110	0.054	0.12	2.71E-04
	40	0	102	0	0.16	6.77E-05
	30	0	99	0	0.16	6.58E-05

The distribution of the release fractions according to the described calculations is shown in Figure 3. The graphs are based on the results of approximately 10000 Monte-Carlo simulations for scenarios with complete fire exposure as well as a randomized exposure between 30 and 100 %. For each scenario the respective fit is also shown.

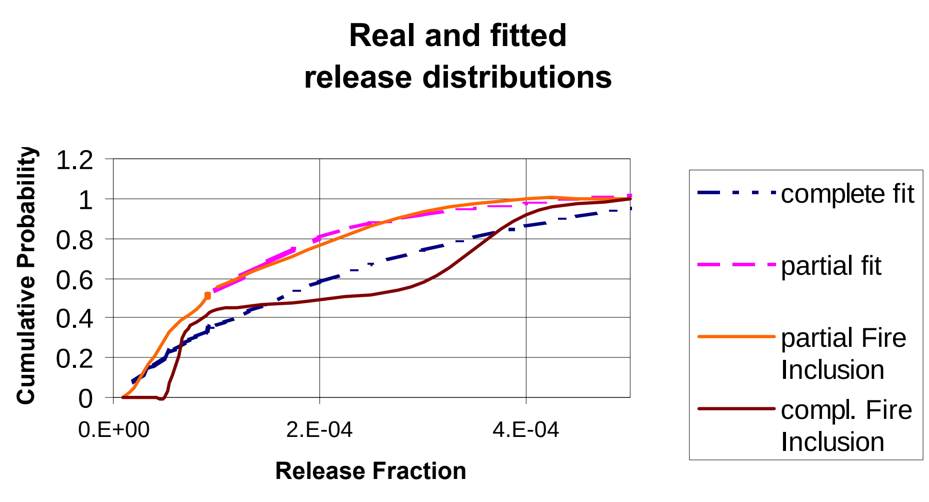

Figure 3: Distributions of release fractions of cemented waste in a fire accident

Similarly, the release fraction distributions are calculated for the following waste products and packages:

1. cemented waste in cylindrical concrete containers
2. bituminized waste in sheet metal containers
3. burnable wastes with low melting points in sheet metal containers
4. burnable wastes in sheet metal containers
5. metallic wastes and scrap in sheet metal containers
6. metallic wastes and scrap in concrete containers
7. unfixed non burnable wastes in sheet metal containers

8. compacted metallic wastes in sheet metal containers
9. compacted metallic wastes in concrete containers
10. compacted non metallic wastes in sheet metal containers
11. compacted non metallic wastes in concrete containers

Mechanical impacts are also considered in addition to thermal load incidents. By so doing, the whole spectrum of low and intermediate level radioactive wastes is covered and the basis for a PSA for disposal facilities prepared.

APPLICATION WITHIN THE FRAMEWORK OF A PSA FOR DISPOSAL FACILITIES

Prior to the PSA the radiological impacts are calculated with Monte-Carlo-simulations based on the distributions of the release fractions for the respective load cases and waste streams. The frequency of the respective incidents and percentage of the waste streams which must be taken into consideration have to be determined.

Incidents, which have to be taken into account for a PSA for a final repository, are

- fire
- mechanical impact with a specific energy < 0.01 J/g
- mechanical impact with a specific energy < 0.03 J/g
- mechanical impact with a specific energy < 0.05 J/g

In the example in figure 4, the fire incident has a probability of occurrence of 30 %. Similarly, the percentage of each waste stream is determined from the total amount of waste.

The procedure of assessing incident doses by the use of Monte-Carlo-simulations is schematically shown in Figure 4. In a first step the scenario and the waste stream are determined by “rolling dice” using random -processes leads to an exact representation of the incident and waste spectra.

For each distinct incident scenario and waste stream a release fraction distribution exists. From this a probabilistic release fraction is determined by the use of a random number.

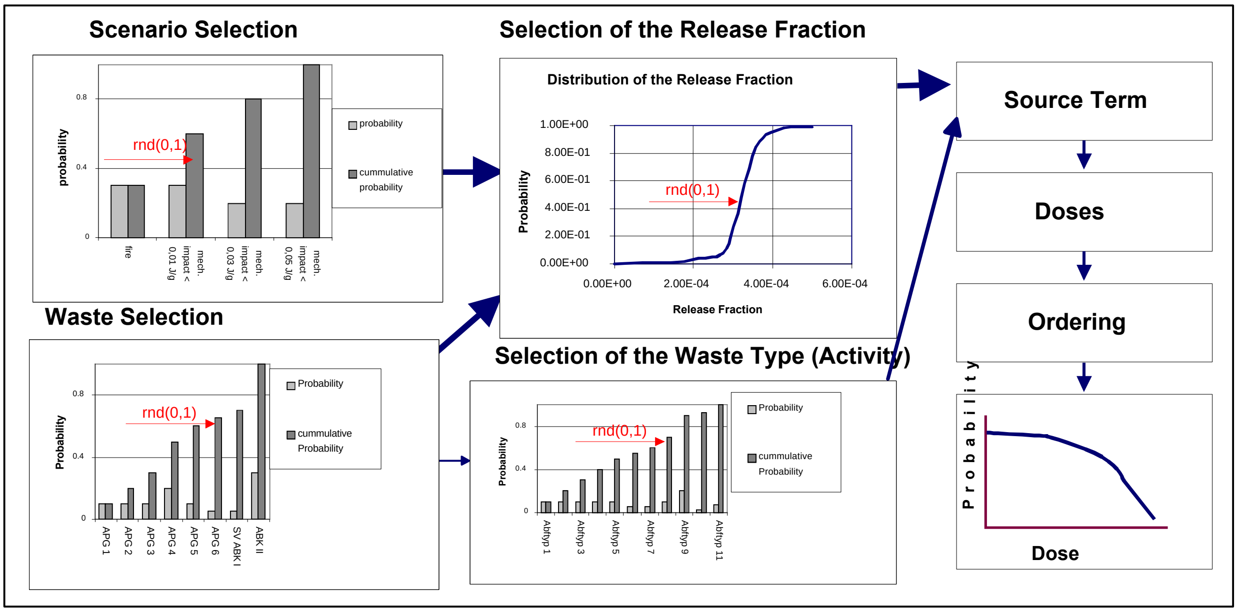

Fig. 4: Schematic of the probabilistic source term determination in a PSA

There is also an activity distribution by waste type for each waste stream as shown in figure 4. The basis for a dose calculation is provided by the waste package activity distribution and the release fraction. After a large number of calculations a dose distribution results which is in exact accordance with

1. the incident distribution,
2. the waste structure,
3. the distribution of the activity inventory in distinct waste streams and
4. the probabilistic distribution of the release fraction.

The dose distribution is represented as the Cumulative Complementary Detriment Function (CCDF). The CCDF provides direct information on the percentage of incidents which stay below given dose limits. Thus, the probabilistic source term determination provides a convenient tool for safety assessment.

Initial results from German LAW and ILLW show that the average dose determined from a probabilistic analysis differs from that of a deterministic source term determination by one to two orders of magnitude.

CONCLUSION

PSA represent a useful tool in the nuclear industry to prove the safety of a facility. Up to now, (particularly in Germany), they have been of minor importance in the field of waste management. In a first feasibility study we came to the conclusion that the extensive tools for PSA that had been developed for nuclear power plants could not be applied to final repositories of radioactive waste. Also to avoid extensive studies as are required for NPPs, we developed a simple approach on the basis of a probabilistic assessment.

The major benefit of our study is to demonstrate the safe operation of a final repository for radioactive waste and the quantification of the conservatism of the deterministic approach. As a consequence this could lead to a relaxation of restrictive waste acceptance criteria without compromising the high safety level of current waste management practice.

Apart from aspects of final disposal our approach can open a new view on the safety of all waste management activities such as treatment, storage and transportation. For example, using the probabilistic approach in determining the consequences of transportation accidents with waste packages it is easily verified that the radiological risk is negligible.

Thus, the probabilistic approach can help in leading to a more objective discussion of the nuclear safety aspects regarding waste management.

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