ACTIVITY RELEASE FROM WASTE PACKAGES CONTAINING LL AND IL WASTE FORMS UNDER MECHANICAL AND THERMAL STRESSES

P. Johnsen, P. Vejmelka, R. Koster
Kernforschungszentrum Karlsruhe GmbH. (KfKO
Institut fur Nukleare Entsorgungstechnik
D-7500 Karlsruhe, FRG

H. Brunner
NUKEM GmbH
D-6450 Hanau, FRG

ABSTRACT

For transport and handling of radioactive waste packages in an underground repository safety assessments are being performed to keep any unacceptable radiation hazards from the operational staff and the population in the site neighborhood. Therefore experiments were carried out to determine source terms for activity release from waste packages containing cemented waste forms in case of heavy mechanical and thermal impacts. Mechanical impact was applied by drop test with a maximum energy input of 3.10^5 Nm. A special cage construction around the target (reinforced concrete covered by a 80 mm steel plate) allows the collection of the airborne fines with a particle size of <10 μm by using micro filters in a defined geometry. In addition, in two experiments the particle fraction with an aerodynamic diameter between 1 μm and 20 μm was determined using a cascade impactor. Additional laboratory experiments were performed to determine comparative values for different waste forms. In case of thermal impact, the temperature profiles in the waste forms were measured and the release of added indicators (Cs, Sr, Eu) was determined. Further laboratory experiments were performed with inactive samples to determine the temperature dependence of water release (Thermogravimetric-Analysis).

INTRODUCTION

For transport and handling of radioactive waste packages in an underground repository safety assessments are performed to keep any unacceptable radiation hazards from the operational staff and the population in the neighborhood of the repository. For this reason it is necessary to know the activity release from waste packages under accidental conditions. In principle an activity release from waste package is possible in case of a thermal or mechanical impact.

Normal transport of radioactive waste packages are goverened by the IAEA-transport regulations [1] which are applied in all cases of transport by land, water or air. At the present state of designing an underground repository in the FRG some impacts on waste packages in a repository are in discussion resulting in higher impacts than those covered by the IAEA-transport regulations. For this reason, experiments were performed to determine source terms for activity release from waste packages containing cemented waste forms.

MECHANICAL IMPACT TESTS

Mechanical impacts on waste packages during the operational phase of the underground repository in principle may cause release of small, contaminated particles (fines) which may be dispersed by the ventilation and thus create an airborne hazard. No quantitative investigations on this problem have been performed with full scale packages. In the USA laboratory experiments with small, unpacked samples were performed to determine the relationship between the mechanical impact energy input and the surface area increase [2,3]. The results of these experiments are useful to characterize waste forms and to compare different waste forms, but it is not possible to extrapolate to the release behavior of full scale waste packages. Drop tests using inactive simulated full-scale packages with cemented waste forms were performed in Finland [4]. For the experiments a fall height of 20-m was applied, but only the disintegration of the packages was documented. Experiments were started to determine quantitatively the amount and the particle size distribution of the dust released from packaged inactive simulated cemented waste forms by mechanical impact. A point of special importance was the determination of the release of fines with a particle size <10 μm because the particles could be inhaled and thus create internal radiation exposure. In addition, laboratory experiments were performed for comparison of the release behavior of different waste forms. The composition of the waste forms is given in Table I.

Laboratory Experiments

The results of laboratory experiments are useful to characterize waste forms and to compare different waste types, but they overestimate airborne release because they neglect the influence of packaging. Some similar laboratory tests have been performed within our investigations too, the results are reported here.

The impact resistance of small laboratory samples was investigated similar to Wallace and Kelly [2]. In the test prismatic samples 2X2X4 cm were impacted and crushed by a falling 1 kg-weight from 1-m height (10 impacts). The impact energy was 2,6 J/g. Particle size distributions were analyzed by sieve analysis according to DIN 4188 and Laserdiffraction techniques for particle sizes smaller than 50 μm.

Weight fractions of fine particles $\leq$ 200 μm for different waste types, surface area increase rates and ratios of surface area increase/impact energy are summarized in Table II. Cement products with compressive strengths $\geq$ 20 N/mm² yield weight fractions up to 5.8 weight % for particles $\leq$ 200 μm and up to 0.91 weight % for particles $\leq$ 50 μm. The average surface area increase yields a factor of 40 compared to the non crushed product, the average ratio of surface area increase/impact energy 20 cm²/J. The results clearly

TABLE I

Composition of Different Cemented Waste Forms

Waste Types	Binder	Waste Loading	w/c-ratio
evaporator concentrate ($NaNO_3$)	BFS-cement OPC	10 wght %	0.35 - 0.45
decontaminations * effluents	BFS-cement	2 wght %	0.50
Scrub water (NaCl) *	BFS-cement	4 wght %	0.40
Filter aid sludge	BFS-cement	7 wght %	0.50
bead resins	BFS-cement	10 wght %	0.28 - 0.33**
incinerator ash	BFS-cement	40-61 wght %	0.40 - 0.50
pyrolysis ashed	OPC	39 wght %	1.80
pyrolysis ashes	BFS-Cement	39 wght %	1.80
compacted trash ***	OPC-mortar	50-82 wght %	0.30 - 0.40

* used for cementation of incinerator ash
** free water
*** pellets Ø 500 mm, overpoured with mortar

TABLE II

Particle size distribution of fines after mechanical impact on cemented laboratory samples (impact energy 2.6 Joule/g)

Waste Types	Weight Fractions ≤ 11 µm	≤ 50 µm	≤ 80 µm	≤ 200 µm	Surface Area Increase (-)	Surface Area/ Impact Energy (cm²/J)
evapor. conc.	2.3 E-3	9.0 E-3	-	3.4 E-2	44	21
decontamination effluents *	1.5 E-3	6.6 E-3	1.9 E-2	4.6 E-2	42	18
filter aid sludge	1.2 E-3	7.1 E-3	1.2 E-2	2.7 E-2	37	20
bead resins	2.1 E-3	9.1 E-3	2.4 E-2	5.8 E-2	47	28
incinerator ash	1.4 E-3	3.4 E-3	1.2 E-2	3.0 E-2	40	16
pyrolysis ashes						
OPC **		4.8 E-3	3.6 E-2	1.4 E-1	58	43
BFS-cement ***		2.8 E-3	1.9 E-2	5.7 E-2	44	30

* with 61 weight % incinerator ash
** compressive strength 4 N/mm²
*** compressive strength 20 N/mm²

Surface area of broken waste product:

$$A = \frac{6M}{S} \sum_{i=1}^{n} \frac{f_i}{\bar{X}_i}$$

A = Surface area of the product (m²)

M = total mass of the particles (kg)

S = density of the waste product (kg/m³)

fi = *weight fraction of class i*

$\bar{x}_i$ = average diameter of particles in class i (m) indicate increasing fine particle formation with decreasing compressive strength.

Full Scale Experiments

Description of the Experimental Setup

From our preliminary experiments it was known that dust release from waste packages due to mechanical impact depends essentially on the container stability. For the drop tests inactive simulated cemented waste forms (OPC 35, w/c = 0.35, 10 weight % $NaNO_3$) packed in 200 l steel drums were used.

The mechanical impact was applied by dropping the waste package from a defined height on to a reinforced concrete target (2x2x3 m, 20 tons) covered by a 80-mm thick steel plate. Different mechanical impacts could be realized using a crane with a maximum lift height of 65-m. Based on the package weight of about 500 kg and the applied drop height of 60-m the mechanical impact yields 3.10^5 Nm. This impact corresponds to the maximum mechanical impact considered for the safety assessments for the repository.

To establish a closed system for the collection of the released dust, (under undisturbed conditions) the concrete basement was surrounded by a mechanically stable cage with mobile ceiling. For collecting a part of the airborne fines, a total of 30 filter units with microfilters (nucleopore 0.2 μm) were placed on the inner sides of the cage. The experimental equipment is shown in Fig. 1. The arrangement of the filter units was selected in accordance with the Institut of Filter- and Aerosoltechnik (LAF) of the Kernforschungszentrum Karlsruhe. From the amount of dust on the filters and taking into account the suck-off parameters, the dust concentration in the cage air volume and in consequence the source term can be calculated.

For the determination of the dust amount on the filter SEM-photographs of selected filter sections were performed and evaluated using an automatic picture evaluation system (Zeiss Company). With this system it was possible to count the total number of particles and to classify them into fractions with different particle size. In addition, the time dependence of the average dust concentration in the cage was measured by using a tyndallometer equipment. To obtain further information on package behavior and dust spreading the impact on the waste package was observed by an installed video camera. In two experiments, the fraction with an aerodynamic diameter between 1 μm and 20 μm was determined using a cascade impactor.

Performance of the Experiments and Results

From preliminary experiments it was known that the largest destruction of the package occurs if the impact is on the drum side. Therefore, only this drop position was applied. Thirdly seconds after the package impact when all the coarse dust has already settled the vacuum pumps of the filter equipment were started and the microfilters were loaded over defined time periods with fine dust and at the same time the tyndallometer measurements were started.

The dust concentration in the air of the cavity and the particle size distribution of this fraction were determined by analyses of the filter sections. The result from four similar experiments show that the release of fines with a particle size <10 μm amounts to $\leq$ 1 g. This value corresponds to 10^{-4}% of the total inventory. The released fraction with an aerodynamic diameter between 1μm and 20μm (impactor measurements) amounts to 1.5 g. These values and the dust concentration measured with the tyndallometer equipment show a good agreement within the range of the same order of magnitude. From these results it is concluded that for homogeneous cement

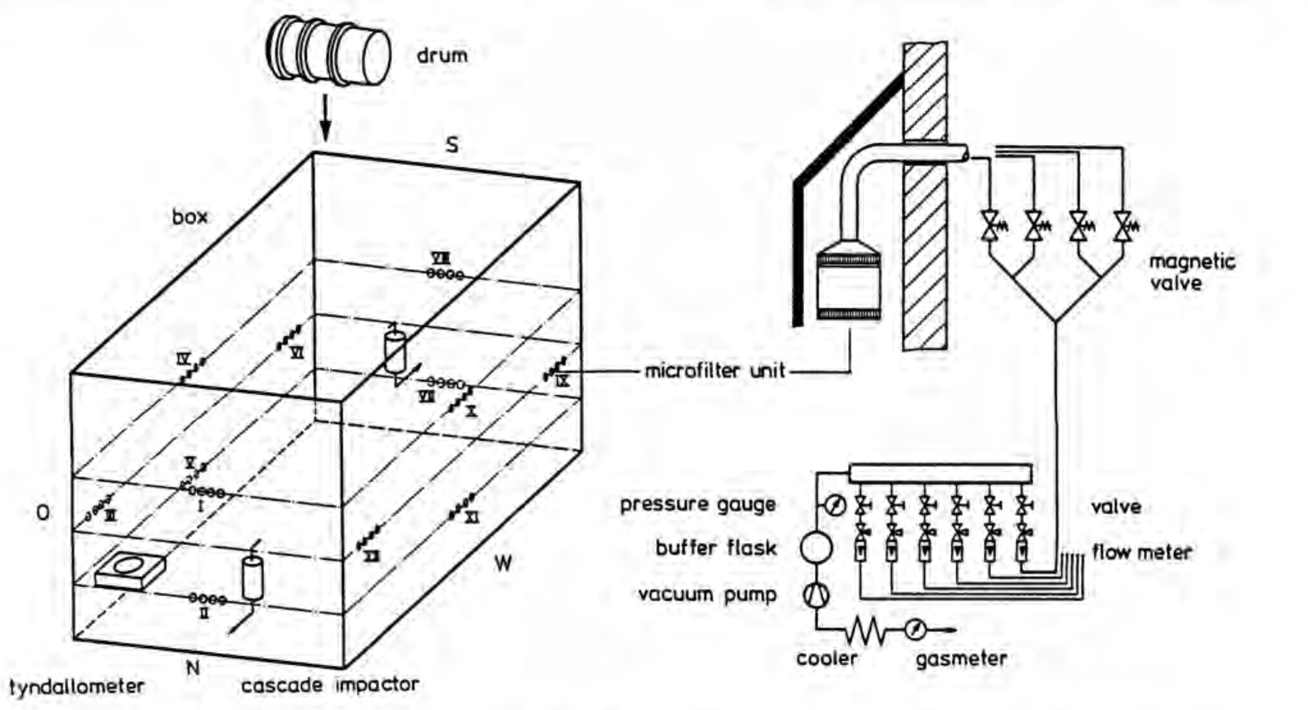

Fig. 1. Scheme of the experimental set up for the collection of airborne fines from the waste package after mechanical impact.

waste forms the activity release amounts also to 10^{-4}% of the radionuclide inventory.

To obtain also information about the waste form destruction at the end of the experiment all the crushed waste form material released from the package was collected for further analyses. The total material weight was determined and a sieve analysis was performed. By the applied separation technique (sieve analysis) only the total fraction with a particle size 0.063 mm could be determined. For a more detailed analysis of the particle size distribution in this range other separation techniques have to be used. The total fraction with a particle size <0.125 mm amounts to 518-608 g, the fraction <0.063 mm amounts to 86-202 g.

THERMAL IMPACT TESTS

Cement as an inorganic binding material is not inflammable, but cementitious waste forms contain a certain amount of water, which can be removed by heating, and in addition organic materials, e.g. ion exchange materials or burnable trash, which can be decomposed by pyrolysis. The release of water or pyrolysis products from cemented waste forms can cause activity release, e.g. by formation of radioactive aerosols.

Hydrated cement contains in principle two different forms of water. The free or unbound water is easily removed from the cement by heating to a temperature of 105°C. The other form is bound in the various hydrated phases and as interlayer water. Release of this water depends on the decomposition points of the hydrated phases and therefore strongly on the temperature. For this reason water release from a cemented waste form in case of an accidental fire situation clearly depends on the energy input.

Experimental

To determine the activity release from cemented waste forms containing $NaNO_3$, laboratory experiments with inactive simulated as well as experiments with inactive simulated full scale samples were performed.

Thermographimetric-Analysis and Differential-Thermo-Analysis Investigations were performed with inactive simulated laboratory samples. Most of the obtained results are already described in Ref. 5 and 6, but for a complete set of data it seems advantageous to perform these experiments.

Samples of different compositions were investigated, that is cement without $NaNO_3$ and cement containing 10 weight % $NaNO_3$. Figure 2 shows the calculated weight losses from the Thermographic-Curve for the two different sample compositions. The results indicate very clearly that most of the weight loss occurs in the temperature range up to 150°C - 200°C. Free pore water is liberated up to a temperature of 105°C and amounts to one third of the total weight loss for this sample composition. An *interesting* point is the observed weight loss for the $NaNO_3$-containing sample in the temperature range between 500°C and 700°C. This increase of weight loss is probably due to decomposition of nitrates and this is also indicated by the total weight loss of 31% because the water content of these samples is only 25.7%.

Field Tests With 200 l and 400 l Packages

In the field tests 200 l- and 400 l- steel drums containing simulated cemented waste forms (evaporator concentrate, filter aid sludges, incinerator ashscrubwater, organic bead resins, high force compacted trash) were exposed to an oil fire or isopropanol fire of 30-110 min duration (flame temperature 800-1100°C). During these experiments the temperature distributions inside the drums, weight losses and element losses (Cs, Sr, Eu, I) were measured.

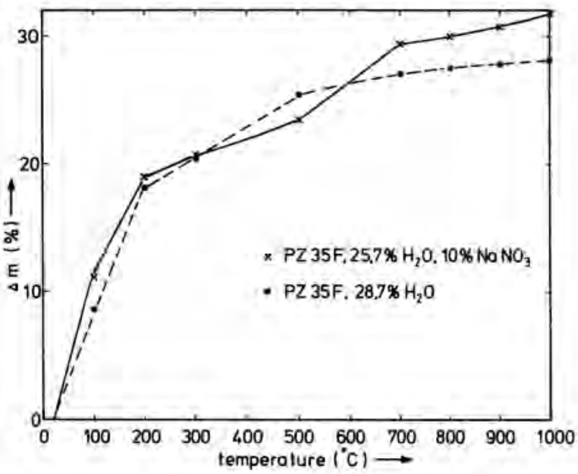

Fig. 2. Weight loss of cemented waste forms (laboratory scale) as function of temperature.

Some results of detailed temperature profile measurements in a homogeneous waste form (evaporator concentrate) are shown in Fig. 3. The maximum temperature at the package surface is 500-660°C. In the waste form a strong temperature gradient exists:

- max. 10 mm from the outer surface temperatures 350°C.
- max. 50 mm from the outer surface temperatures 100°C.
- max. temperature inside the waste form 80°C 10 hours after the end of the fire.

This result indicates very clearly that water and pyrolysis residues are released only from the outer parts of the waste form. In case of heterogeneous waste forms, e.g. compacted burnable trash in pellets, overpoured with mortar (10 mm mortar thickness) higher temperatures inside the pellets up to 400°C can occur, due to the different heat capacity of the heterogeneous structure and pyrolysis reaction inside the waste.

Weight losses of 200 l drums are 10 - 16 kg after 30 min. fire and 16 -23 kg after the 60 min. fire test. This corresponds to 2.3 - 4.0 weight % and 4.0-7.3 weight % respectively. The highest weight losses has been obtained with compacted burnable trash (23 kg/7.3 weight %). Fire tests with 400 l-samples (evaporator concentrates) yielded weight losses of 14-15 kg (1.8-2.0 weight %) after a 30 min. fire test.

The element losses were investigated by two different offgas sampling methods:

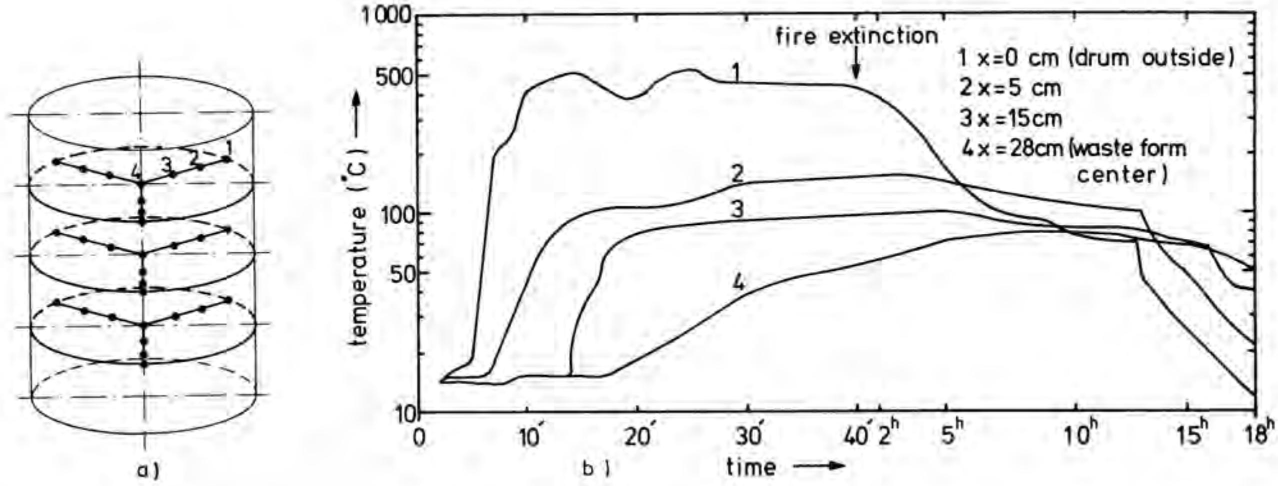

Fig. 3. Positions of the thermocouples in the cemented waste form (a) and time dependence of the temperature distribution in the waste form (upper level of thermocouples) during the fire test (field test, 40 min. oil fire).

- total condensation
- total exhaustion and aerosol filtration (Fig. 4).

Cs, Sr, Eu and I-releases from different cementitious waste forms after a 60 min. fire test (200 l drums) are summarized in Table III. Homogeneous cement products yield following results:

Total released fractions

Cs $\leq$ 3. E-4
Sr $\leq$ 9. E-5
I $\leq$ 2. E-4
Eu $\leq$ 9. E-6.

High force compacted burnable trash shows higher release rates, due to higher product temperatures inside the waste:

Total released fractions

Cs 2. E-3
Sr 1. E-4

The data apply to cement products solidified without supernatant water in open steel drums without additional shielding.

Volume changes and water losses during heating cause some crack formation at the surface of homogeneous cement products. An outer layer of 5 mm product shows color changes, indicating high temperatures and pyrolysis effects. During heating of overpoured compacted burnable trash a volume increase of pellets occurred (2-3 Vol.%) which caused partial destruction of the mortar layer surrounding the waste. Inside the pellets an outer layer of 5-10 mm was thermally decomposed.

In summary the results of the experiments give a clear indication that any activity release from cemented waste forms under thermal impact is very low. For example, the activity release from $NaNO_3$-containing cemented waste forms is 4 orders of magnitude lower than the activity release from $NaNO_3$-containing bituminized waste forms [7].

REFERENCES

1. Regulations for the Safe Transport of Radioactive Materials. 1973 Revised Edition Safety Series No. 6 IAEA, Vienna (1979)

2. R.M. WALLACE, J.A. KELLY. An Impact Test for Solid Waste Forms, DP-1400 (1976)

3. H.K. MANAKTALA, P. COLOMBO, R.M. NEILSON. Properties of Radioactive Waste Containers, Quarterly Progress Report Oct.-Dec. 1979, BNL-NUREG-51148, Feb. 1980

4. E. RÄMO, U. PULKONEN, J.P. AITOLLA. Damage of Reactor Waste Packages in Transport Accidents. NKA (AO) (79) 17 (1979)

5. W. MANNS. Über den Wassergehalt von Beton bei höheren Temperaturen. Betontechnische Berichte 17 (1975).

6. R.O. LOKKEN. A Review of Radioactive Waste Immobilization in Concrete. PNL-2654 UC-70 (June 1978).

7. W. KLUGER, P. VEJMELKA, R. KÖSTER. Investigation of Activity Release from Bituminized Intermediate Level Waste Forms under Thermal Stresses. IAEA-Symposium on the Conditioning of Radioactive Wastes for Storage and Disposal, Utrecht, Netherland (1982).

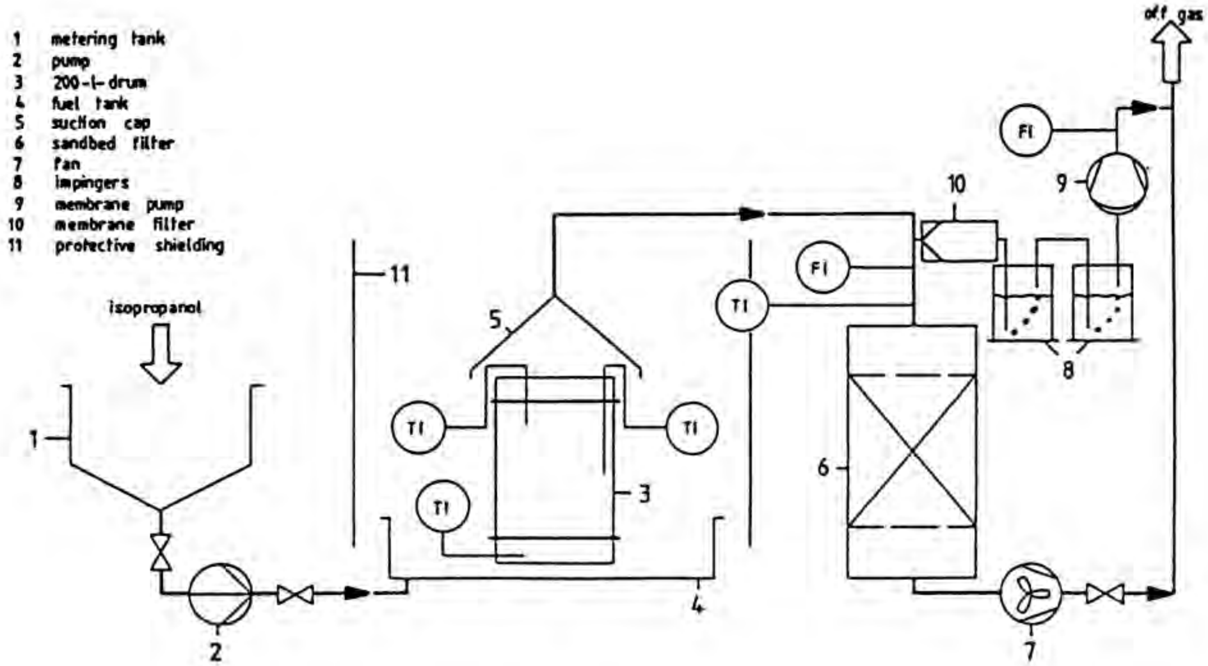

Fig. 4. Flow sheet of experimental set up for the fire field test.

TABLE III

Element-specific release fractions from different waste forms after 60 min. fire test, 200 l-drums *

Cemented Waste Form	Release Fractions (-)			
	Cs	Sr	Eu	I
Evaporator conc.	8 E-5	2 E-5	4 E-6	-
Bead resins	8 E-5	-	5 E-6	2 E-4
Incinerator ash/ scrub water	3 E-4	9 E-5	9 E-6	-
Compacted Trash	2 E-3	1 E-4	-	-

* without lid