RELEASE OF RADIOACTIVITY IN FIRE ACCIDENTS

W. U. Boetsch, D. Gründler
Institute for Safety Technology (ISTec) GmbH
Schwertnergasse 1,
D-50667 Cologne, Germany

J. Thiel
Federal Office for Radiation Protection (BfS)
P. O. Box 10 01 49
D-38201 Salzgitter

ABSTRACT

The operational safety of a repository for radioactive waste is mainly achieved with the control of the risk of fire accidents. Various experiments were performed to target at the adjustment of theoretical models within safety and risk analysis of such installation. The investigations included the study of the release mechanisms caused by combustion of radioactive waste and the melting of radioactive metallic scrap as well as the research of the behavior of an inactive waste drum in an open fire and the release ratio out of waste simulation material. Due to the results of this project corrections of the classification of waste groups could be made and safety calculations for fire accidents could be renewed and become more realistic.

INTRODUCTION

Fire accidents are of highest significance for the operational safety of a repository for radioactive wastes. The design of waste packages must ensure that in case of fires dose limits in the environment are not exceeded. In order to proof a sufficient safety level the release behaviour of the waste product as well as the waste package in case of fire has to be known. Up to now many investigations were carried out with regard to the determination of release fraction in case of thermal impact. However, the results of these investigations are suitable only for particular wastes and boundary conditions and could not be used for a general approach.

In order to develop a comprehensive approach for the determination of release fractions under fire conditions a broad research project has been carried out in Germany on behalf of BfS (Federal Office for Radiation Protection). The aim of this project has been to provide parameter to be used in theoretical models which have been developed within the framework of safety analysis for interim storage and disposal. Parameters of interest are heat transfer parameters and nuclide specific temperature depended release rates for various wastes.

The research project consists of four elements:

1. Investigation of the behaviour of drums in a fire
   (Validation of computer codes and the derivation of heat transfer parameter).
2. Analysis of the release mechanism "combustion" (feed, off-gas and ashes).

3. Investigation of radioactive releases from metallic scrap under thermal exposure.
4. Inactive investigations of model samples for several waste characteristics (cemented waste, concentrates, mixed waste, metallic waste, resins and combustible waste).

The results obtained with these topics are shown in the following.

Validation of Fire Simulating Computer Codes

Experimental heating-up of typical waste drums in a fire under various fire scenarios has been performed at the iBMB (Institut für Baustoffe, Massivbau und Brandschutz) of Braunschweig University of Technology, Germany. For prediction of the potential consequences of fires these pool fire experiments were important for the validation of the available fire models and codes for this application.

For the experiments a large oven with various openings was used. Ten different scenarios were carried out. Both the fire room and the drum were instrumented with thermocouples.

Test Specification

Within the pool fire test series the fire compartment OSKAR of iBMB with a floor size of 3.6 m x 3.6 m and a height of 5.7 m has been used [1]. This facility has 3 possible openings for the natural ventilation of the fire room. At the ceiling the hot gases and smoke can be extracted and cleaned by a fan system with filters. During the experiments gas and surface temperatures, gas composition, velocities and heat flux densities have been measured.

Description of the Test Facility

The test facility has a floor area of 3.6 m x 3.6 m and a height of 5.7 m. In the center of the floor area a pan with a size of 4 m^2 has been installed on a weight scale. The bottom level of this pan has an elevation of 0.3 m. The side walls are approx. 0.4 m high.

A photo of the view through the front door (interior view) is shown in Fig. 1.

Fig. 1 View through the front door

Targets

Behind the large pan a typical barrel type waste drum has been installed. The center of the barrel bottom is located at (x = 1.8m, y = 3.2 m, z = 0.6 m). This container has a diameter of 0.64 m and a height of 0.96 m. It is a double vessel container. The diameter of the inner barrel is 0.515 m. The inner barrel consists of tinplate and is filled with styrene divinylbenzene copolymer with sulfur acid groups (ion exchanger granulates). The gap between the inner and outer steel barrel has been filled with concrete. Table I gives the properties of the used materials.

Table I Properties of the target materials

Material	**Heat conductivity λ [W/mK]**	**Heat capacity c_p [J/kgK]**	**Density ρ [kg/m^3]**
Granulate (styrene)	0.233	1600	1000
Tinplate	63	230	7280
Concrete (between barrels)	2.1	880	2400
Steel	44.5	480	7743
Concrete (probe)	2.1	880	2400
Aerated concrete	0.11	1350	420

Performed Measurements

For measuring the temperatures inside the fire compartment 3 mm thick thermocouples have been used. These were not protected against flame radiation. The position of the thermocouples has been fixed with a grid. Table II lists the performed measurements.

Along three lines the temperatures inside the barrel container were measured (M36 to M53). The velocity inside the plume, at the door and inside the fan systems was measured by bi-directional probes.

The cross section area at the measurement positions V11 and V12 was 0.4 m x 0.8 m and at the velocity measurement position V13 inside the hood the diameter of the pipe is 0.4 m.

To measure the gas concentrations and pressure open pipes are routed to the outside of the fire compartment and connected to the measurement systems.

Table II List of performed measurements

Nomenclature WS = Heat flow density measurement M = Temperature measurement GA = Measurement of gas composition V = Gas velocity measurement P = Measurement of total pressure GV = Measurement of weight loss
Measurement position
Plume-temperature (x = 175, y = 195)**:**
M 1 – M 6 (z = 100 cm – z = 520)
Temperature inside fire compartment:
Level 1: (z = 150 cm)
M 7 (x = 275; y = 85) – M 10 (x = 90; y = 280)
Level 2: (z = 335 cm)
M 11 (x = 275; y = 85) – M 14 (x = 90; y = 280)
Level 3: (z = 520 cm)
M 15 (x = 275; y = 85) – M 18 (x = 90; y = 280)
Surface temperature
Plates on the surface
M 19 (x = 245; y = 360; z = 150) - M 21 (x = 0; y = 190; z = 170)
"Coated thermocouples" on the surface:
M 22 (x = 245; y = 360; z = 150) - M 24 (x = 0; y = 190; z = 170)
Fire load temperature:
M25 (x = 175; y = 195; z = 33)
Barrel type target (waste package):
upper level (z = 140)
M36 (x = 180; y = 288; z = 140) - M41 (x = 180; y = 320; z = 140)
center level (z = 110)
M42 (x = 180; y = 288; z = 110) - M47 (x = 180; y = 320; z = 110)
lower level (z = 85)
M48 (x = 180; y = 288; z = 85) - M53 (x = 180; y = 320; z = 85)
Heat flow density:
WS 1 (x = 360; y = 150; z = 180) - WS 4 (x = 0; y = 70; z = 170)
Weight loss of fire load:
GV 1 (x = 180; y = 180; z = 0)

The temperature gradient in a typical waste drum caused by a nearby pool fire is shown in Fig. 2.

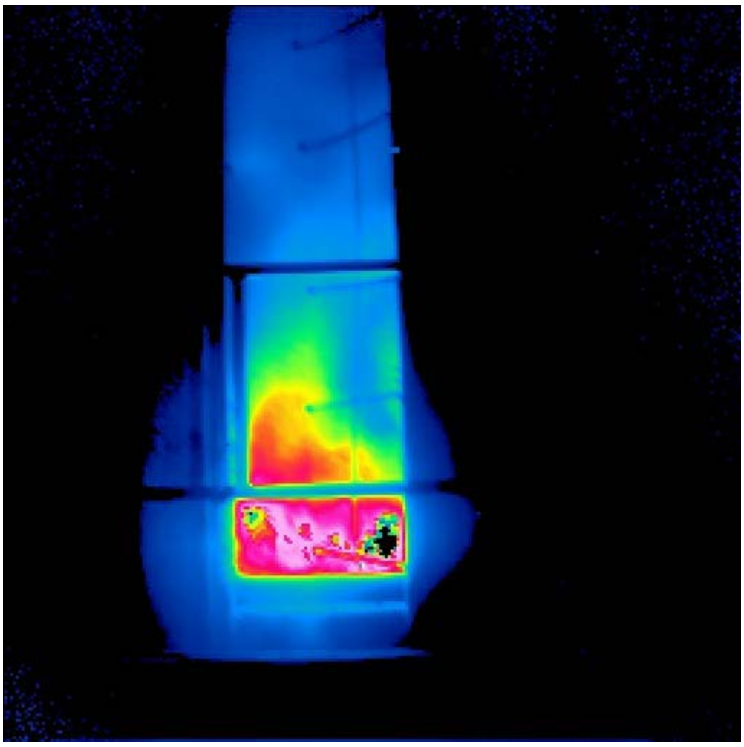

Fig. 2 Temperature gradient during an open pool fire

Results

These experiments described above led to applicative results which allowed the validation of the computer codes CFAST and HEATING (7.1) as well as the derivation of heat transfer parameter.

Release Rates of Incinerated Radioactive Wastes

Focus of the examinations of balances of an industrial incineration plant was the analysis of the release mechanism "combustion". In order to gain representative results the release behaviour of the combustion plant of the Jülich research centre has been studied. The nuclide specific release fractions are derived from activity balances (feed, off-gas and ashes).

Facility Description

The Jülich JÜV-50/2 incinerator at the FZ-Jülich Research Center, Germany, is designed for the incineration of solid and liquid radioactive waste [2].

The furnace is divided in the following sections from top to bottom:

- charging station with glove box
- lock chamber
- gasification chamber

- primary burner
- ash funnel

The gasification chamber is separated from the primary burner by a pair of flaps and the primary burner from the ash funnel by a movable ash grate.

Operation of the JÜV-50/2

The generally untreated solid waste collected in 200l drums and filter boxes in one piece are passed through the glove box and lock chamber into the gasification chamber preheated to ignition temperature. Then they are partly burned and gasified at approx. 800° C by adding a substoichiometric amount of atmospheric oxygen.

The incineration/ gasification conditions in the gasification chamber are controlled via a controlled air section in which fresh air and moist off-gas are adequately mixed taking the temperature at the outlet of the primary burner as basis. Sudden power variations due to excessive heating value fluctuations of the solid waste to be burned are controlled by injecting steam into the gasification chamber.

Below the pair of flaps the primary burner is located into which primary air is blown through the flaps. The temperature in this chamber is 850 °C -1100 °C. The off-gas is passed into the lateral secondary burner. The ash drops onto the ash grate supported by the poking movements of the flaps where it burns out before being discharged through the cooling funnel.

Incineration takes place at constant temperature and produces very low CO values. The dust discharge is relatively small due to the flow conditions. The secondary burner is designed for the residence time of two seconds at 1200 °C. Operation takes place at a temperature reduced to $T \geq 850$ °C.

Burnable radioactive liquid waste is fed into the secondary burner and/or the primary burner through different lances and burned. The hot off-gases are cooled directly behind the secondary burner by quench and dust and part of the pollutant gases are washed out in the acid and neutral stages of a two-stage scrubber. The wash water is filtered and passed to the waste water treatment line for vaporization.

The precleaned cooled off-gas is then passed in a moisture-saturated state into a wet activated carbon column where dioxins and other hazardous substances are absorbed.

The last stage of gas purification consists of two parallel high efficiency particulate absorber (HEPA) filters. Prior to entering these filters the off-gas is electrically heated to avoid the temperature falling below the dew point in the filters.

The incinerator's nominal throughput is 50kg/h based on a heat content of 20.000 KJ/kg of waste. The annual throughput is limited by the license to 140 Mg/a (120Mg/a of solid waste and 20 Mg/a of liquid waste).

Release Balances and Results

The JÜV 50/2 incinerator is a commercially operated facility. Therefore waste treatment is given by economy and operational logistic as well as by legal requirements such as Federal Immission Control Ordinance or German Radiation Protection Ordinance. The investigation of release ratios of combusted waste depends on clear balancing of the in- and output amounts of one incineration cycle. Unfortunately the analysis of radioactive releases in the off-gas and ashes appeared to be practicable in only very few cases because of cross contamination effects and impurities during the campaign wise operation. As far as balances could be drawn the release ratios former meet theoretical assumptions in comparable dimensions.

Releases from Metallic Scrap

The release fraction for metallic scrap has been analyzed in technical scale at the Siempelkamp melting plant. The samples are packed in drums, which are heated in an inductive melting oven. The studies were carried out for pure black and white steel as well as for zinc-coated and varnished metals. The samples are representative materials from nuclear power plants.

The CARLA Melting Plant

In October 1989 the **C**entrale **A**nlage zum **R**ezyklieren **L**eichtaktiver **A**bfälle started operation. Core of the plant is a 3,2 t medium frequency induction furnace, located in an inner housing [3]. Primarily the furnace is being operated for melting steel scrap, but replacement by a special crucible allows melting of non-ferrous metal, too. The inner housing is the barrier to the cutting and sorting hall for preparing the melting charges. The CARLA plant is licensed by the German Radiation Protection Ordinance.

Experimental set-up

A total of 7 experiments were carried out with several representative sample materials, as given in Table III [4]. Each sample composition was placed in a common 200 l steel drum.

Table III Sample compositions

Nr.	Sample material
BV1	Fe- lacquer coated
BV2	Fe- zinc coated; (shear scrap)
BV3	Vanadium Steel; (shear scrap)
BV4	Fe- coated; (shear scrap, compressed)
BV5	Fe- zinc coated; (shear scrap)
BV6	Fe- zinc coated; (shear scrap)
BV7	Fe- zinc coated; (shear scrap)

The average weight of the sample drums was 310 kg. The sample materials was "real" radioactive waste which had to be packed into the 200 l drums to ensure the purity of each sample charge and to install the thermocouples in a well defined 3-dimensional arrangement. Vent openings were cut into the head of the drum to allow controlled air circulation of the drum content.

The thermocouples were led through the top cover. An exhaust pipe connected the top cover with the plenum chamber (Fig.3). Top cover and drum weren't screwed tightly to prevent possible explosions in consequence of pressure build-up in the drum.

Each drum was equipped with 8 thermocouples for temperature documentation reasons, which continuous data flow was collected by a data logging station. All data were displayed on a computer system simultaneously as well.

During the entire experiments (heat-up and dwell time) off-gas samples were collected constantly. The off-gas was led through a filter station equipped with changeable glass fiber filters. About 20 filters per experiment were collected. After weighing the filters to achieve balance data concerning the mass release they were analyzed radiological (gamma-spectrometric, detection time 3600 s, nuclide specific).

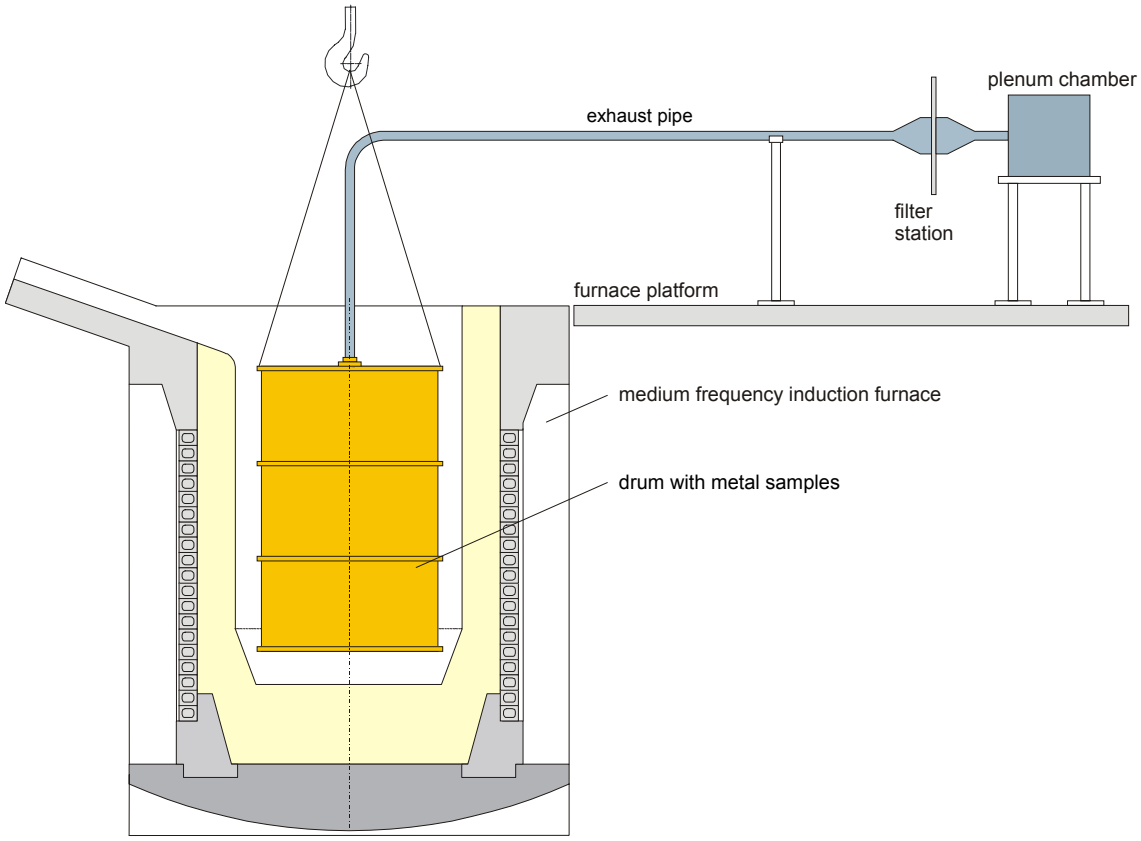

Fig. 3 Experimental set-up

Experimental Sequence

The experiments were carried out subsequent to a standard melting process to gain from the residual heat. After all measuring equipment was installed the furnace was heated up inductively. Regarding to the filling level and sample material the examined drums were heated up to 800 ° C for one hour and the nuclide specific release rates are measured.

Release Quantifications and Results

Prior to and after the execution of the experiments the nuclide specific activities were determined accurately. Table IV gives a list of integral released radioactivity for Ag-110m, Co-60 and Cs-137.

Table IV Integral released radioactivity for Ag-110m, Co-60 and Cs-137

No.	Radioactivity (Input)			Radioactivity (off-gas)			Radioactivity (release)		
	Ag-110m	Co-60	Cs-137	Ag-110m	Co-60	Cs-137	Ag-110m	Co-60	Cs-137
	[Bq]	[Bq]	[Bq]	[Bq]	[Bq]	[Bq]	[%]	[%]	[%]
BV1	--	5,330E+05	1,290E+05	--	1,385E+01	3,383E+03	--	0,003	2,622
BV2	--	5,000E+05	1,210E+05	--	5,445E+00	1,957E+02	--	0,001	0,162
BV3	--	2,200E+05	1,290E+05	--	1,503E+01	2,456E+01	--	0,007	0,019
BV4	--	6,220E+05	7,380E+04	--	7,138E+00	1,387E+01	--	0,001	0,019
BV5	--	6,760E+05	8,020E+04	--	3,978E+00	1,223E+01	--	0,001	0,015
BV6	1,186E+05	2,845E+04	1,524E+03	2,023E+01	4,099E+00	8,694E+00	0,017	0,014	0,370
BV7	1,046E+05	5,862E+05	7,676E+03	2,790E+01	7,402E+00	2,068E+01	0,027	0,001	0,269

For the calculation of the release ratios in case of a fire only release mechanisms like sublimation or vaporization were considered. These calculated ratios match very well with the results of the executed experiments at the CARLA melting plant. The calculations were made for safety considerations of waste containments for disposal at the (planned) German disposal at the former iron ore mine KONRAD. The hypothetical factors of the safety analysis were approved as well.

Inactive investigations of model samples

Another experimental sequence of this research project is the investigation of inactive samples which represent all main waste streams. One declared aim is to complete the knowledge of the properties of these inactive elements which occur in chemical and physical compounds similar to real radioactive waste. In a next step the transferability of these results to the circumstances of real waste has to be checked.

Sample characterization

These samples simulated various waste products such as cemented waste, concentrates, mixed waste, metallic waste, resins and combustible waste. All samples were precisely doped with the following different elements:

Li, Na, K, Cs, Be, Mg, Ca, Sr, Ba, Al - Sn, Pb, P, As, Sb, Bi, S, Se, Cu, Zn, Cd, Sc, Y, Ti, Zr, V, Cr, Mo, Mn, Fe, Co, Ni, Ru, Pd, Ce, Sm, Eu, Th

The purpose of this selection was to cover the most from the periodic table of the elements.

Sample Preparation

The samples were doped with certain amounts of inactive elements mentioned above. As representatives for the main waste streams the following materials were used as carriers:

I. Cemented waste:
   a) inactive elements applied dissolved in solution, solution is used to prepare cement sample
   b) nactive elements applied on dry cement sample
II. Mixed waste:
   Representative mixture from rubber, paper, textile etc. (Fig. 4) doped with dissolved elements
III. Metallic waste:
   Steel wool is sprayed with element solution and dried before testing
IV. Concentrates:
   a) m-boric acid doped with dissolved elements
   b) m-boric acid mixed with doped and dried cellulose
V. Ion exchange granulates:
   Common ion exchange granulates are doped with solution from dissolved elements

Fig. 4 Mixed Waste Material

Experimental Set-up

All release investigations with inactive samples were executed at the chemical laboratory of ANALYTIS in Wesseling, Germany. Most of the needed instruments belong to the standard

equipment of an analytical laboratory. Only the reactor had to be constructed in respect of the required gas tightness for a quantitative gas determination.

After preparing the sample and placing it in the gastight reactor both was heated up to distinct temperatures to measure the specific release rates of the doped elements out of each carrier material. Therefore the temperature was increased and held in the steps 200°C, 400 °C, 600 °C and 800°C for one hour. A stream of Argon was led through the reactor to assure the quantitative rate of yield of released elements at each temperature step. All released gas was led through acid and alkaline solutions. The washing solutions were changed every new temperature step and transferred to the quantitative analysis unit to determine the amount of released material at a distinct temperature. Analyzing units were ICP-OES (Inductively Coupled Plasma Optical Emission Spectroscopy), ICP-MS (Inductively Coupled Plasma Mass Spectroscopy) and AAS (Atom Absorption Spectroscopy).

Results

Due to the fact that the analysis was executed for each single element being doped on carrier material and that for each examined temperature step 3 washing solutions needed to be analyzed a very big amount of values resulted from this experiment sequence. In some cases the build-up of the achieved release rate of an element (for instance Sb on a concrete carrier) wasn't plausible because of non-typical release behaviour at lower temperatures. The development of Sb release doesn't seem to follow known physical principles. One explanation for this could be found in the application circumstances of the doped elements. The elements were applied in mixed salt solutions with several different anions as counter ions. Salt recombination due to increasing temperature and processes related to the dynamics of chemical equilibration might be the origin for such implausibility. The transferability of the results of this experimental sequence is therefore restricted to more or less general additive information about the physical release behaviour of elements and less suitable for the direct application in nuclear concerns. The applicability of the results can be examined due to the correlation from release values with temperature distribution in a heated waste containment to allow the calculation of release rates of waste products in waste containers.

CONCLUSION

The presented sequence of experiments was performed to complete the knowledge of the release behavior of radioactive nuclides in the case of fire and to verify and validate existing theoretical models or computer systems. Regarding to long term safety considerations of a disposal for radioactive wastes or the qualification of waste containments it was necessary to identify conservative assumptions or models to prevent avoidable uncertainties. Due to the results of the investigation of metal scrap for instance a correction of the classification of waste groups could be made. Adequately safety calculations for fire accidents can be renewed and become now more realistic.

REFERENCES

1 D. Hosser, R. Dobbernack, "Untersuchungen zum Erwärmungsverhalten eines Gebindes bei Treibstofflachenbränden", Juni 2003, iBMB TU Braunschweig

2 S. Halaszovich, Personal Information about the JÜV 50/2 incinerator, Dec. 2003.

3 U. Quade, "Radiological Characterization of Steel Scrap Recycling by Melting", Siempelkamp Nukleartechnik GmbH, Krefeld, Germany, 2001

4 N. Dyllong, "Versuche zur Untersuchung von Freisetzung aus metallischen Abfällen im thermischen Lastfall", Siempelkamp Arbeitsbericht, SNT-2003-02, July 2003.