

EXPERIENCES WITH THE RELEASE OF RADIOACTIVE WASTES AND PROBLEMS RELATED TO THEIR STORAGE IN A FINAL REPOSITORY

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ABSTRACT

First, this paper describes an approach of waste disposal for naturally occurring radioactive materials (NORM) from non-nuclear industries. Secondly, a limited amount of waste resulting from core melt ex-vessel tests, contaminated with depleted uranium, is to be disposed of by shallow land burial under the condition of free release. An exposure scenario via ground water yields an individual dose rate of $< 10 \mu\text{Sv/a}$. Based on these results, slag from melting fuel contaminated scrap should also be managed in the same way, because after special treatment, the activity in the slag is comparable to this in the slag from the test melt.

INTRODUCTION

Waste treatment, disposal or release is fully regulated in the nuclear industry. In non-nuclear industry (NNI), like oil & gas production, fertilizer, mineral sand processing and steel industry, large amounts of waste with naturally occurring radioactivity (NORM) are produced and have to be handled in an ecological and economic manner. To harmonize the regulations in the EC, the member states have induced the European Commission to issue a revised Council Directive, containing the new Basic Safety Standard (BSS) ¹. The BSS contains exemption levels of radioactive materials specified for each single radionuclide. These exemption levels for natural radionuclides are substantially lower than the former value of 500 Bq/g in general for NORM. Consequently, large amounts of waste coming from the NNI have to be declared „radioactive“ in the future, when these clearance levels will be applied. The disposal of the waste could involve substantial financial, practical and logistic problems. On the other hand, waste, resulted from melting of NORM contaminated scrap from the natural gas extraction industry has not been accepted in the German final depository Morsleben, as the extremely low limit for α - emitters was exceeded. The conflict reflects the demand for a practicable solution.

In this context, two further waste disposal problems are under discussion with German authorities.

In the frame of European research activities on nuclear safety, experimental work on ex-vessel core melt behavior has been performed at Siempelkamp. Several core melting tests have been performed using depleted uranium from fuel element production. From these melting tests, approx. 70 t of waste have to be disposed of. Studies investigated for disposal of the waste on shallow land burial result in an annual dose of $< 10 \mu\text{Sv/h}$, which is within the “de minimis” philosophy of the IAEA.

Slag resulting from melting metallic waste coming from dismantling fuel fabrication plants has to be treated in a special manner. That means, the enriched uranium contamination of the scrap is mixed by adding depleted uranium to the melt, to yield a natural uranium matrix in the slag. Now the option should be investigated to dispose of the slag also in shallow land burial when the “de minimis” concept is fulfilled.

MELTING NORM - CONTAMINATED STEEL SCRAP

Siempelkamp is operating two plants for melting radioactively contaminated scrap. CARLA plant is licensed to accept material which exceeds the exemption level up to a factor of 2, which means

200 Bq/g for material from the nuclear industry
1000 Bq/g for material from the NNI.

The GERTA plant², especially established to melt chemically- and/or NORM - contaminated scrap is limited to handle NORM - contaminated scrap with less than 500 Bq/g (exemption level for NORM). A lot of material from the oil & gas industry is contaminated by a mixture of NORM and mercury and, therefore, has to be treated in the GERTA plant. 350 t of pipes, valves, heat exchangers, etc. from site remediation of former east Germany natural gas extraction, containing scale activity in the range of 250 - 480 Bq/g and an average mercury content of 8 mg/kg have been cleaned by melting. Mechanisms in the gas extraction process itself cause a separation of the daughters from the mother nuclide. In the uranium - radium decay chain only nuclides down from Ra 226 and in the thorium decay chain down from Ra 228 have been screened in the scale analysis³.

Nuclide behavior by melting is quite different. Ra 226 resp. Ra 228 and some of their daughters are transferred into the slag, while Pb 210 evaporates at the melting temperature of steel at 1,300 °C and passes the exhaust system. Screening of filter dust samples shows a peak at 46,5 keV γ -line from Pb 210. The measurable residual activity in the cast ingots is well below 1 Bq/g. The melting campaign of 350 t scrap waste results in 18 t of slag with an average specific activity of 93 Bq/g (Ra 226: 57 Bq/g); approx. 1 t of filter dust with an average activity of 535 Bq/g and 3.6 t of floor sweepings with an average activity of 255 Bq/g have to be disposed of. 4 waste drums exceeded the exemption level of 500 Bq/g. The federal collection depot for radioactive waste offered a storage for 3 drums to a price of 475,000 DM, the fourth drum was refused, because the Ra 226 activity could not be accepted.

This situation reduced the recycling of NORM contaminated scrap to absurdity for the industry and investigations have been started to find a practicable and economic waste management.

Brenk Systemplanung, a consultant of the German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety was appointed to evaluate the radiological impact of waste streams like

- recycling to road construction material
- shallow land burial
- sidewalk construction
- playground
- parking lot ⁴.

Taking into account a maximum expected slag of 100 t/a with an average Ra 226 activity of 65 Bq/g, the resulting individual dose from external γ - radiation is shown in Table I.

Table I: Dose calculation for external γ - radiation from 100 t/a NOR contaminated slag

parameter	scenario				
	road construction	con-shallow land burial	sidewalk	playground	parking lot
dose rate [(μ Sv/h)/(Bq _{Ra226} /g _{slag})]	0.54	0.00078	0.54	0.45 ⁽¹⁾	0.11
mass specific Ra 226 activity [Bq/g]	65	65	65	65	65
mixture rate	no	--	0.05	0.05	0.05
residence time [h/a]	8 - 20	1800	50 - 100	200 - 450	1000 - 1800
dose [μ Sv/a]	280 - 700	90	90 - 180	290 - 660	360 - 640

(1) 1 m above an infinite area with 5 cm thickness and a density of 1.8 t/m³

In each scenario, the annual dose is below 1 mSv, which is tolerable for NORM sources. Based on these results, Siempelkamp was licensed to recycle up to 100 t slag with a max. Ra 226 activity of 65 Bq/g mixed together with slag coming from the own foundry by a minimum mixing rate of a factor 4. The license is limited until implementation of the BSS in the German radiation protection ordinance.

WASTE FROM CORE MELT EXPERIMENTS

In the frame of European research activities on nuclear safety, experimental work on ex-vessel core melt behavior has been performed at Siempelkamp ⁵. Approx. 3 t corium, melted in the CARLA furnace have been spread out on various material surfaces, to study their behavior and to verify computer codes. To simulate the fuel in the corium, depleted UO₂ - powder was added to

the melt. From seven experimental set-ups, a total 70 t of waste contaminated by depleted uranium have to disposed of.

Table II lists the nuclide specific activity in the waste.

Table II: Specific uranium activities and relating mass for the COMAS test set-up

Test set – up	U 238 [Bq]	U 234 [Bq]	U 235 [Bq]	mass [kg]
COMAS 1 - R	6.65×10^9	1.85×10^9	8.31×10^7	11,636
COMAS 5	8.75×10^9	1.53×10^9	9.11×10^7	11,993
COMAS 5 a	3.35×10^9	2.12×10^9	9.49×10^7	5,742
COMAS 6	6.62×10^9	3.35×10^9	1.34×10^7	8,771
COMAS EU - 4	5.69×10^9	3.58×10^9	1.61×10^7	11,619
COMAS EU - 1		inactive test		6,140
COMAS EU - 2		inactive test		13,812
total activity [Bq]	3.11×10^{10}	1.24×10^{10}	5.64×10^8	69,713
spec. Act. [Bq/g]	445.5	178.0	8.1	

The German final repository at Morsleben (now closed by court decision) was not licensed to accept the waste due to the strong limitation of U 238 to 2.2×10^4 Bq/m³ equivalent to 4.4×10^{-2} Bq/g (density of 500 kg/m³ assumed). Under these circumstances, the licensing board decided to recycle most of the material in planned further experiments and licensed Siempelkamp an intermediate storage for the waste on site up to the end of the year 2002.

Nevertheless, Siempelkamp is in discussion with the authorities to find an alternative disposal solution. One of them is shallow land burial on the condition that the waste is accorded free release. First investigations proved the radiation exposure of this scenario and showed the result to reflect the “de minimis” concept. A crucial part is the long-term assessment of prognostic public exposure via groundwater. Migration processes influence the radionuclide concentration. After leaching the total activity of COMAS waste in some thousand years, spring water activity will be approx. 0.09 Bq-U238/l, which is in the range of measured geogene uranium concentrations in the drinking water in some east German regions today. The exposure via ingestion by using the spring water results in only 9 µSv/a individual dose in some thousand years⁶. It could be shown that under conservative assumptions the radiation exposure to the public is in the range of 10 µSv/a. Second step was to find a depository which is willing to accept the waste. A high standard depository for industrial hazardous waste in Northrhine-Westphalia will accept the waste fixed in concrete blocks, provided that the waste is accorded free release by the authorities. These results give Siempelkamp the opportunity to apply for free release for the COMAS waste. An answer is expected in the first half of 1999.

SLAG WITH URANIUM IN A NATURAL MATRIX

From decommissioning of Siemens fuel element fabrication plant at Hanau, a lot of scrap with contamination of enriched uranium is delivered to Siempelkamp for melting in the CARLA plant under controlled conditions. The contract covers a mass of 2,000 t of scrap. From the melting process, a max. amount of 200 t slag is expected over a period of two years. The German radiation protection ordinance limits the fuel content in waste to 15 g/100 kg (in the past 3 g/100 kg). Due to the enrichment of the uranium activity in the slag, the limit can be exceeded. To avoid this, Siempelkamp is licensed to add depleted uranium to the melt/slag to such an extent that uranium in natural distribution - U 235 (0,72 %) and U 238 (99,28 %) - is formed in the slag. This slag is comparable to waste from the core melt tests described above. Table III compares the specific activities.

Table III: Specific uranium activities in COMAS waste and Siemens slag

isotope	COMAS - waste [Bq/g]	Siemens slag [Bq/g]	clearance for shallow land burial [Bq/g]
U 234	178	210	9
U 235	8	10	3
U 238	445	520	10

Based on the results in ⁶ and taking into account an annual amount of 100 t slag, an individual dose in the same range resulting for the scenario of COMAS waste depository can be expected. When the “de minimis” concept is fulfilled, free release for disposal of the slag on a high standard depository for industrial hazardous waste will be applied for.

CONCLUSIONS

Waste resulting from melting of scrap from the oil- and gas industry contaminated by NORM can be recycled to road construction material. The dose exposure is less than 1 mSv/a for a mass of 100 t/a and a limit of 65 Bq/g Ra 226 in the waste.

For waste depository scenarios from core melt experiments and comparable slag from melting scrap from decommissioning of fuel element fabrication, individual doses in the range of 10 μ Sv/a have been calculated.

The investigations show alternative waste management solutions for limited amount of special waste. Case by case licensing is necessary.

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