

RELEASE OF RESIDUES FROM MELTING NORM-CONTAMINATED STEEL SCRAP

- A German Approach -

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ABSTRACT

As many raw materials like crude oil, natural gas, mineral sands, phosphore ores and others are contaminated by radionuclides from the Uranium and/or Thorium decay chain (NORM), also plants for processing these materials became contaminated during operation. When plants are shut down, large quantities of pipes, valves, pumps and other components have to be scrapped. As scrap yards and steel mills are equipped by large detector systems to avoid an input of radioactivity into the steel cycle, decontamination is required before recycling.

Siempelkamp is operating a melting plant for processing NORM and/or chemically/ toxically contaminated steel scrap. Beside the decontaminated steel as output, residues like slag and filter dust have to be managed within the range of licensed values. Based on the European Safety Standard the European member states have to implement radiation exposure from work activities with NORM in their Radiation Protection Ordinances (RPO). The German government revised the RPO in July 2001. Part 3 describes exposure limits for workers and for the public. Exposures from residues management have to meet 1 mSv/year.

Brenk Systemplanung has performed calculations for assessing the radiation exposure from residues of the Siempelkamp melting plant. These calculations have been based on the input of metal from different origins and include all relevant exposure pathways in a number of scenarios. The calculations have been based on the dose criterion of 1 mSv/y as required by the German RPO. The methods and results will be presented.

INTRODUCTION

Even in non-nuclear industries like oil & gas extraction, fertilizer production or paper mills, plant systems can become contaminated by plate outs from radionuclides of the U238 and Th232 decay chains.

In case of scrapping components, scrap yards will detect the scrap as radioactive by large entrance detector systems. To recover the steel and to minimize the waste a predecontamination by melting is a proven option. As Siempelkamp has been melting radioactive materials from nuclear facilities since 1989 up to a total amount of 15,000 t, the company has already references to also melt scrap contaminated with naturally occurring radionuclides (NORM).

In 1998, the melting plant GERTA started operation to process NORM and/or chemically/toxically contaminated steel scrap. This plant is licensed to melt NORM scrap below the exemption level of 500 Bq/g according to the former German Radiation Protection Ordinance.

The German Radiation Protection Ordinance as of July 20, 2001 for the first time contains a systematic framework of regulations protecting workers and the public against radiation exposures from residues of industrial and mining processes containing enhanced levels of naturally occurring radionuclides [1]. These regulations satisfy the requirements of the European Council Directive 96/29/EURATOM (Basic Safety Standards) [2]. The overall goal of the new regulations is to keep the additional effective dose for the population from the recycling and disposal of NORM below 1 mSv/a. A set of exemption criteria in terms of the mass specific activity of the U 238 and Th 232 decay chains in the material has been established for various recycling and disposal options. If these values are not exceeded, the material may be recycled or disposed of without restrictions. If, however, this set of exemption criteria is not applicable to a certain material (e.g. because only very small masses arise annually, or because a specific disposal or recycling option is chosen which is not covered), there still remains the possibility of a case specific evaluation of the radiological consequences. In the present case, a case specific assessment of slags from the GERTA plant was performed for various disposal and recycling options because only small masses (in the range of 100 Mg/a) are involved.

THE MELTING PLANT GERTA

Since 1989, Siempelkamp is operating a melting plant for radioactively contaminated scrap resulting from the nuclear sector [3]. This plant, which is run at the headquarters in Krefeld, Germany, is one of the three relevant melting plants in Europe. 15,000 t of scrap from nuclear power plant operation and decommissioning projects have already been successfully melted and in a wide range recycled to products which can be re-used in the nuclear industry. By this process high volume could be saved for intermediate and final waste storage. In the early 90^{ies}, this service was requested from non-nuclear industries, like crude oil and natural gas extraction, which required an ecological recycling concept for NORM scrap. Scrap from these branches is often cross-contaminated by toxic chemicals like mercury, PCB, dioxine/furane, which require special filters in the off-gas system of the furnace used. As the off-gas system in the first melting plant could not be improved, a second melting plant for the treatment of NORM-scrap and/or chemical/toxical contamination started operation in December 1998 [4]. The plant is licensed under the federal law on protection against environmental pollution (BImSchG). To avoid licensing under the atomic law, the specific activity of NORM had to be limited to 500 Bq/g. Up

to now, some 5,000 t of this kind of scrap have been successfully processed, 1,300 t of which were NORM-contaminated.

The recycling process is based on a net frequency induction furnace with a capacity of 8 t. All incoming material has to pass through the dismantling area to be cut to chargeable sizes. Three bunkers are available to store material enough for a two-week melting run, which allows separation of materials from different customers or different kinds of contamination.

Scrap leaves the bunker and enters the charging device which automatically rolls through a gate to its end position above the furnace. A close connection between the charging device itself and the furnace lid enables off-gas to be extracted to the filter system, which consists of four steps - a cyclone, a cooler, a bag filter and a fixed-bed absorber. Coarse dust is separated in the cyclone and fine dust in the bag filter. The fixed-bed absorber filled with activated carbon is doped with sulphur and serves to separate mercury from the off-gas. In the cooler, the off-gas is cooled to secure optimum bag filter efficiency.

This combination of filters prevents the mercury concentration in the chimney stack from exceeding $50 \mu\text{g}/\text{Nm}^3$, equivalent to only a quarter of the permitted value.

As the NORM-scrap is processed, vaporising radionuclides - especially Pb-210/Po-210 - condense and are absorbed by the dust and retained in the cyclone and bag filter.

The melting process in the 8 t net frequency induction furnace starts up with a 3 t liquid iron basic melt followed by a 5-t-batch of scrap. The furnace can be operated in temperatures ranging from 1,350 to 1,500°C, although a special furnace lining is required with the upper temperatures.

The liquid iron is poured into steel moulds resulting in ingots of 1 t each. These ingots are free from any further contamination and can be released for recycling in the iron and steel industry, whereas the slag and filter dust has to be managed considering the requirements from the German radiation protection ordinance.

RADIOLOGICAL ASSESSMENT

Requirements of the German Radiation Protection Ordinance

In its Part 3, the new Radiation Protection Ordinance (RPO) 0 which has entered into force on August 1, 2001, has transformed Title VII of the EURATOM BSS 0 into detailed national legislation. Part 3 (§§ 93 to 104) of the RPO refers to

- NORM at workplaces (§§ 95 to 96),
- the protection of the public from NORM (§§ 97 to 102),
- cosmic radiation in conjunction with flight crews (§ 103).

- For the recycling of NORM the following exposure scenarios were considered:
 - for workers: road construction, use as aggregate for building materials, storage in a warehouse, use for sand-blasting
 - for other members of the general public: living near a road constructed using NORM, living near a public place covered with NORM, living in a house which has been constructed using NORM.
- For the disposal of NORM three exposure scenarios were considered:
 - for workers: disposal on a landfill, disposal in underground mines;
 - for other members of the general public: living near a landfill or a waste rock dump where NORM is being or has been disposed of.

For these scenarios the following exposure pathways have been included: external radiation, inhalation of contaminated dust, inhalation of radon/radon daughter nuclides and direct ingestion of fine grained radioactive material. Where appropriate, also the following exposure pathways were considered: drinking contaminated water (after groundwater migration of contaminants); use of this water for irrigation of agricultural products or for watering cattle (after groundwater migration of contaminants); contamination of agricultural products through the deposition of dust.

For the exemption/clearance levels presented in Annex XII of the German Radiation Protection Ordinance 0, a tiered approach is chosen. That means that multiple values are provided which depend on the material type and the disposal or recycling option. The criteria are expressed in the following form

$$C_{U238max} + C_{Th232max} \leq C, \quad (\text{Eq. 1})$$

where $C_{U238max}$ and $C_{Th232max}$ stand for the highest specific activities of the nuclides in the U 238 and Th 232 decay chains.

1. The primary exemption level for the recycling or disposal of NORM corresponds to an activity concentration **C=1 Bq/g** in equation (1).
2. An exemption/clearance level of **C = 0.5 Bq/g** in equation (1) is applied in cases where more than 5000 Mg/a of residues are disposed of in the vicinity of a usable aquifer, when building materials for the construction of houses contain more than 20 % residues or when building materials for other purposes (road construction, construction of dams etc.) contain more than 50 % residues.
3. For the recycling or disposal of residues in underground mines a higher exemption level of **C = 5 Bq/g** has been defined.

4. For waste rock resulting from mining activities, a lower clearance/exemption level has to be applied if the recycling or disposal of the material leads to the covering of an area of more than 1 ha in the vicinity of a usable aquifer. In this case, the following exemption/clearance levels apply: $C_{U238max} \leq 0,2 \text{ Bq/g}$ and $C_{Th232max} \leq 0,2 \text{ Bq/g}$.
5. A number of further criteria exist which cannot be discussed here.

Suitability of the Requirements of the RPO for Slags from the GERTA Plant

The values listed in the previous section are not suitable for slags from the GERTA plant due to a number of reasons. The main reason is that the masses involved here (100 Mg/a) are by order of magnitudes smaller than those used for the derivation of the set of exemption/clearance levels in Annex XII of the German RPO. It is obvious that small amounts of material which constitute only an insignificant part of the recycling or disposal material stream do not have to be treated on the same basis as e.g. heaps of several 100.000 Mg or even millions of tons.

For this reason, a case specific radiological assessment valuation is necessary. § 98 No. 1 RPO stipulates that such a case specific assessment of the possible doses from recycling or disposal of NORM has to be performed using a dose criterion of 1 mSv/a. The results of that assessment in terms of the mass specific activity will then replace the exemption/clearance levels of Annex XII RPO 0.

Scenarios for Slags from the GERTA Plant

A case specific assessment for the slags of the GERTA plant has been performed on the basis of the following enveloping assumptions:

- The total annual mass of the slag bearing NORM from contaminated scrap will not exceed the range of 100 Mg/a, resulting from 1000 Mg/a of scrap (775 Mg oil and gas industry, 75 Mg/a fertilizer production, 75 Mg/a paper production, 75 Mg/a material with Thorium).
- The slag is removed from the SNT site together with other slags on a regular basis with lorries. The slags are fetched weekly or even several times per week. The assumption of a monthly removal is therefore an enveloping approach.
- The slag quantity from the GERTA plant which is assumed to remain together in a single load is 8 Mg (100 Mg/a divided by 12 removals per year). The material is mixed with other slags with a mixing ratio of at least 0.5. Both assumptions have a conservative nature.

The slags which are collected by a contracting company are currently only used for recycling in road construction. In spite of this, the scenarios cover a wide range of disposal or recycling possibilities because slags may in general be used for a number of options and § 98 No. 1 RPO

stipulates that no restrictions after release of the material shall have to apply. According to an expertise provided by independent experts it is, however, not possible to recycle the slag from the GERTA plant in concrete. This is the reason why no recycling scenarios for use in building construction have been included. The list of scenarios comprises the following:

A. Scenarios for workers:

- A1: milling and sorting of the material;
- A2: use of the material as a cover for public places, without cover;
- A3: use of the material in road construction, covered;
- A4: disposal on a public landfill.

B. Scenarios for other members of the general public:

- B1: stay on public places (without cover);
- B2: stay on public places (with cover);
- B3: dwelling near a landfill site.

All scenarios include dose contributions from external irradiation, inadvertent direct ingestion of material (except for B2 and B3), and inhalation of dust (except for B2). Secondary ingestion pathways via groundwater migration are included for B3. A comparison with the list in section 0 shows that the relevant scenarios used for the derivation of exemption/clearance levels for NORM are covered. The scenarios B1 to B3 are calculated for all 6 age groups as defined by ICRP.

Parameter values cannot be provided here in detail. As far as possible, standard values have been used. Exposure times, dust concentrations etc. have been adjusted to be compatible with the material quantities and the throughput of recycling steps. The resulting activity concentrations, lateral extensions of the areas covered with the slags etc. are calculated in accordance with slag quantities and its activity concentrations.

Results of the Dose Calculations

Dose calculations are performed on the basis of the scenarios listed in section 0 for a unit activity (1 Bq/g) for the U 238 und Th 232 decay chains (1 Bq/g means that each nuclide of the chains is present with this activity). In addition, the sub-chain Ra 226+ is calculated which comprises the nuclides from Ra 226 down to and including Po 214.

Results from the dose calculations for workers (scenarios A1 to A4) yield dose conversion factors in the range of $1 \cdot 10^{-6}$ to $5 \cdot 10^{-6}$ Sv/a per 1 Bq/g. The leading scenario is A1. For members of the general public, the dose conversion factors are in the range of 10^{-8} up to $1.5 \cdot 10^{-5}$ Sv/a per 1 Bq/g with the leading scenario always being B1. It follows that the doses are generally governed by scenarios for the general public.

For the final dose assessment, the doses have to be calculated on the basis of the actual activities of the slag instead of the unit activity. A reasonable assumption for the annual mean values of the activities is:

- U 238 decay chain: ~ 10 Bq/g (per nuclide of the chain)
- Th 232 decay chain: ~ 11 Bq/g (per nuclide of the chain)

The total dose is then calculated to be ~ 0.3 mSv/a.

Additional calculations have been performed for cases where the slag will originate only from melting campaigns dedicated to scrap from single sources as listed in section 0. Because of the fact that some scenarios for the general public do not depend on the annual average but on the activity of a material quantity which remains together in a single batch, different results are obtained in those cases. If only material with a high Th activity would be melted, the resulting slag might have specific activities in the range of 120 Bq/g per nuclide of the Th 232 decay chain and might lead to doses above 1 mSv/a. It is of course within the discretion of SNT to schedule melting in such a way that the resulting slag would not lead to radiological problems.

In addition, those results can further be used for calculating the mass specific or total activity which may be accepted on the scrap from a single waste producer during a certain time span.

CONCLUSIONS

Based on the experience of melting 15,000 t of contaminated scrap from the nuclear cycle, melting of scrap from branches out of the nuclear field, e. g. oil and gas, fertilizer, paper mills and others are proven at Siempelkamp. Therefore, one purpose furnace for NORM and toxically contaminated residues equipped with a special charging device and filter system is in operation. 5,000 t of contaminated scrap, 1,300 t of which were NORM contaminated, have been successfully treated in the plant up to now. The resulting waste from the process like slag and filter dust is managed in an environmentally protective and safe way.

The radiological assessment of recycling and disposal of the slag from the GERTA plant has shown that this slag will lead to doses only which are significantly below the dose criterion of 1 mSv/a for members of the general public. The results of this assessment allow Siempelkamp to plan acceptance of scrap and melting campaigns more easily and reliably and to be able to demonstrate at any time that the release of the slag is in compliance with the German regulations.

LITERATURE

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