#### **RECYCLING OF URANIUM- AND PLUTONIUM-CONTAMINATED METALS FROM DECOMMISSIONING OF THE HANAU FUEL FABRICATION PLANT**

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#### ABSTRACT

Decommissioning of a nuclear facility comprises not only actual dismantling but also, above all, management of the resulting residual materials and waste. Siemens Decommissioning Projects (DP) in Hanau has been involved in this task since 1995 when the decision was taken to decommission and dismantle the Hanau Fuel Fabrication Plant. Due to the decommissioning, large amounts of contaminated steel scrap have to be managed. The contamination of this metal scrap can be found almost exclusively in the form of surface contamination. Various decontamination technologies are involved, as there are

Blasting
 • Wiping

Often these methods are not sufficient to meet the free release limits. In these cases, SIEMENS has decided to melt the scrap at Siempelkamp's melting plant. The plant is licensed according to the German Radiation Protection Ordinance § 7 (issue of 20.07.2001). The furnace is a medium frequency induction type with a load capacity of 3.2 t and a throughput of 2 t/h for steel melting. For safety reasons, the furnace is widely operated by remote handling. A highly efficient filter system of cyclone, bag filter and HEPA-filter in two lines retains the dust and aerosol activity from the off-gas system. The slag is solidified at the surface of the melt and gripped before pouring the liquid iron into a chill. Since 1989, in total 15,000 t have been molten in the plant, 2,000 t of them having been contaminated steel scrap from the decommissioning of fuel fabrication plants. Decontamination factors could be achieved between 80 and 100 by the high affinity of the uranium to the slag former. The activity is transferred to the slag up to nearly 100 %. Samples taken from metal, slag and dust are analyzed by gamma measurements of the 186 keV line of U235 and the 1001 keV line of Pa234m for U238. All produced ingots showed a remaining activity less than 1 Bq/g and could be released for industrial reuse.

#### **INTRODUCTION**

Decommissioning of a nuclear facility comprises not only actual dismantling but also, above all, management of the resulting residual materials and waste. Siemens Decommissioning Projects (DP) in Hanau has been involved in this task since 1995 when the decision was taken to decommission and dismantle the Hanau Fuel Fabrication Plant.

During planning of the dismantling of the uranium and mixed-oxide (MOX) fuel fabrication facilities, it already became evident that, by the time dismantling was completed, around 3,000 Mg of uranium-contaminated metals and 1,200 Mg of steel scrap slightly contaminated with plutonium would arise in the MOX facility.

Both the German Waste Management Act and the Atomic Energy Act require that priority be given to recycling such materials – insofar as such recycling produces a product suitable for unrestricted reuse and is economically justifiable – in order to reduce the volume of waste requiring disposal. These requirements can only be met through decontamination of the metals at a cost lower than that of radwaste disposal.

The new German Radiological Protection Ordinance of July 2001 defines nuclide-specific clearance levels below which materials can be reused without restrictions. The reuse of metals contaminated with uranium from light water reactor (LWR) fuel assemblies is unrestricted if their residual activity is  $\leq 0.5$  Bq/g. This figure supercedes the clearance level of 0.1 Bq/g previously recommended by the German Radiological Protection Commission (SSK). If the metal is recycled in a steelmaking process, residual uranium activity levels of up to 2 Bq/g are permitted. In the case of plutonium, the clearance level for unrestricted reuse is calculated to be 0.04 Bq/g for the given isotopic mixture and 0.2 Bq/g for metal recycling.

One of the options available for decontaminating such metals is to melt them in a furnace together with slag-forming additives which chemically bind the uranium and plutonium. This process was tested on an industrial scale and optimized in the early 1990s. Optimization was aimed primarily at reducing the volume of radioactive secondary waste – slag, furnace refractory and dust – which must be stored in a final repository and therefore generates additional costs.

Siemens DP in Hanau has developed a technique for disposing of this secondary waste that avoids any additional final storage costs. According to the acceptance criteria for the Konrad repository, waste drums may only be delivered inside cuboid Konrad Containers. To immobilize these drums inside the containers, the internal spaces between the drums must be backfilled with concrete or other materials. The requirements specified for final storage specifically permit radioactive materials to be added to the immobilization materials when Class 1 Konrad Containers are being used. All of the secondary waste produced by the above melting process is mineral in nature and is therefore eminently suitable for use as aggregates in concrete backfill. Depending on the specific Konrad Container design, the waste drums occupy approximately 50 % of the container volume; i.e. for each 200-liter drum there remains a free internal volume of 200 l in which up to 60 l of aggregates can be accommodated in the concrete backfill without requiring additional final storage capacity. For the more than 1000 waste drums arising from the Hanau decommissioning project, this therefore represents an efficient option for disposing of the secondary waste from metal recycling which does not generate any additional costs.

# MELTING OF URANIUM CONTAMINATED STEEL SCRAP

Due to the decommissioning of the uranium and MOX-fuel fabrication plants of Siemens, Hanau, large amounts of contaminated steel scrap have to be managed. The contamination of this metal scrap is almost exclusively in the form of surface contamination. Various decontamination technologies are involved, as there are

• Blasting • Wiping

Often these methods are not sufficient to meet the free release limits. In these cases, SIEMENS has decided to melt the scrap at Siempelkamp's melting plant CARLA. The metal scrap is presorted at Hanau site and placed in 200 l drums. All drums are measured by means of a stationary germanium detector before shipping to the melting plant to evaluate the activity inventory for each nuclide, taking into account the nuclide quota of the fuel fabrication plant.

### THE MELTING PLANT CARLA

The CARLA (<u>C</u>entrale <u>A</u>nlage zum <u>R</u>ecyclieren <u>L</u>eichtaktiver <u>A</u>bfälle) melting plant started operation in 1989, [1]. The radiological input limits of scrap are

200 Bq/g ( $\alpha$ ,  $\beta$ ,  $\gamma$ ) 2,000 Bq/g ( $\Sigma$  H3, C14, Fe55, Ni63) 100 Bq/g ( $\Sigma$  U233, U235, Pu239, Pu241) and < 15 g/100 kg for fissionable nuclides.

The plant is licensed according of the German Radiation Protection Ordinance § 7 (issue of 20.07.2001).

The melting furnace is a medium frequency induction type with a load capacity of 3.2 t and a throughput of 2 t/h for steel melting (ref. Fig. 1). For safety reasons, the furnace is widely operated by remote handling from an operation desk outside the housing where the furnace is located. Only slagging, temperature control and sample taking needs access to the furnace. A highly efficient filter system of cyclone, bag filter and HEPA-filter in two lines retains the dust and aerosol activity from the off-gas system. The slag is solidified at the surface of the melt and gripped before pouring the liquid iron into a chill where ingots of 1 t each are produced.

Since 1989, in total 15,000 t have been molten in the plant, 2,000 t of them having been contaminated steel scrap from the decommissioning of Siemens' and Nukem's fuel fabrication plants at Hanau.

# EXPERIENCES FROM MELTING URANIUM CONTAMINATED SCRAP FROM SIEMENS FUEL FABRICATION

The targets of the treatment by melting were to release the metal after melting and to minimize the secondary waste. First investigations to study the behaviour of uranium and the decontamination efficiency by the melting process have been started in 1992 [2], [3]. Decontamination factors could be achieved between 80 and 100 by the high affinity of the uranium to the slag former. In first test melts, the slag was liquefied and poured into a chill to assure homogenization of the uranium in the slag. As this process resulted in an amount of 15 % of secondary waste in kinds of slag, dust and crucible, the slagging has been improved to minimize secondary waste.

The advantage of using a medium frequency induction furnace as smelter is the efficient mixing of the melt by electromagnetic forces. By the turbulent conditions on the melt surface, the transferred uranium is sufficiently homogenized in the slag to avoid clusters of uranium in the slag. The activity is transferred to the slag up to nearly 100 %. Samples taken from metal, slag and dust are analyzed by gamma measurements of the 186 keV line of U235 and the 1001 keV line of Pa234m for U238.

Until October 2001, about 1,760 t of ingots with remaining activity less than 1 Bq/g could be released for industrial reuse. In the past, 12 % of the ingots had to be treated by a second melt to achieve the release limit.

To avoid this inefficiency, the melt is now controlled by taking samples from the melt after slagging which are quickly measured by gammaspectroscopy to check the efficiency of decontamination during the melting process. If the remaining activity is too high for release, slagging has to be repeated.

The learned lessons finally reduced the amount of secondary waste down to 8 %. In total, 94 t of slag, 30 t of dust and 58 t of crucible have been produced. 27 t of waste could be released for shallow land depository. To save final disposal volume, the slag and crucible is planned to be milled down to sizes less than 2 cm which allows to add this material to concrete for filling space volume in KONRAD containers. First tests of milling the slag show good results. Investigations to press the dust into sizes which meet the criteria for using as additive to filling concrete are going on.

# MELTING OF STEEL SCRAP FROM MOX-FUEL FABRICATION

The decommissioning of the MOX-fuel fabrication plant at Hanau has started in 2001. The scrapped process components will be volume reduced by high pressure and packaged in final disposal containers. For the process-building structures, safe release of materials cannot be assured. An amount of 1,180 t of steel scrap is expected. As release measurements of this scrap are time and cost consuming, the steel scrap should be melted. By an amount of 10 t a test melt will be arranged in early 2002. It is expected that the metal as well as the waste can be released after melting. For the ingots, the release value for recycling of 0.2 Bq/g has to be fulfilled.

# CONCLUSION

Especially for uranium contaminated steel scrap melting is an efficient method for decontamination. The produced ingots could be released for industrial reuse (< 1 Bq/g) in the past. In view of the Radiation Protection Ordinance, issued on 20.07.2001, a remaining activity of 2 Bq/g for uranium as well as 0.2 Bq/g for plutonium has to be undercut. Waste resulting from the melting process can be widely used for space filling of final disposal containers.



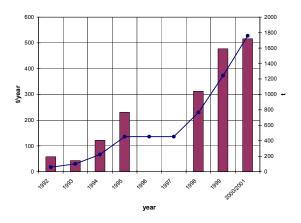


Fig. 2: Molten steel scrap from Siemens Hanau decommissioning projects

Fig. 1: CARLA induction furnace

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