

NON-HAZARDOUS RE-USE OF COMPONENTS
REMOVED FROM NUCLEAR FACILITIES BY
MELTING PROCESSES

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ABSTRACT

Melting processes are used to homogenize and or to decontaminate radioactive contaminated scrap originating from repair, refitting or decommissioning of nuclear facilities. Test melts in the laboratory scale are performed with β , γ -contaminated material in order to decide whether or not components have to be pre-decontaminated before they are melted in the foundry. About 500 t have already been treated in this way for the re-use in nuclear facilities.

Melt experiments in the laboratory scale with α -contaminated materials resulted in a one step compaction and decontamination process. The α -radionuclides are nearly quantitatively released from the melt into the slag. A simple direct α -measuring technique was developed to control melt processes.

INTRODUCTION

The method given priority in the German Atomic Energy Act is to recycle decommissioned metal parts from nuclear facilities.

Various types of contaminated piping, valves, heat exchangers, vessels and steel components accumulate in the course of repairs, refitting or decommissioning of nuclear facilities. Depending on their origin these components are contaminated with various radionuclides. In many cases chemical decontamination of such large components with complex geometric surfaces cannot be performed economically.

For this reason Kraftwerk Union AG (KWU) started in the late seventies with experimental work on the decontamination of metal scrap using melting procedures. The first experimental phase was concentrated on the behavior of β , γ -contaminated material. At present melting procedures for α -contaminated scrap are under investigation.

Melting Procedures in the Laboratory Scale

The first investigations were performed in a small muffle furnace. For several years a Nernst-Tammann type high temperature unit has been in use.

This unit consists of a transformer and a high temperature furnace. The transformer is continuously adjustable and has a power output up to 23 kVA. Thus temperatures up to 1700°C can be reached. The furnace is slewable. Thus metal and slag can be separated by pouring off if necessary. Usually Carborundum tube crucibles are used for the melting experiments. The melting crucible with an inside diameter of 60 mm is located inside a safety crucible with an inside diameter of 105 mm in order to avoid contamination

of the furnace in the event of a crucible failure. The melting crucible can hold 3-5 kg material for each melting experiment.

Above the furnace an adjustable chimney head is installed which is connected by a metal tube to the off-gas system of the control area of the radiochemical laboratory. An additional aerosol filter is installed after the chimney head in order to collect all aerosol activity which may be released from the melt.

The temperature is measured by a Pt-Rh-Pt thermocouple which is covered by a ceramic tube.

The use of high tube crucibles with an overall length of 300 mm allows the production of fairly thin ingots. This has the advantage of easy control of the activity distribution along the axial length of the small ingot.

Melt Experiments with β , γ -Contaminated Materials

The laboratory melting unit is used for pre-tests in the recycling of metallic components from nuclear power plants. These components are β , γ -contaminated. The main radionuclides are Co 60 and Cs 137. The reason for test melts prior to large scale melt downs in the foundry is to decide if the activity concentration of the melt is within the license of the foundry. Furthermore the test melt can answer the question as to whether the large scale melt can be used for products without any restrictions in the release or for products for re-use in nuclear facilities or if a coarse decontamination step has to be performed prior to the melt down in the foundry.

Representative test material is taken from decommissioned components, melted down in the KWU-laboratory unit and the activity concentration in the small ingot is measured. The experience gained from a lot of test melts shows that the residual activity in the metallic phase correlates well to the residual activity measured after the large scale melt down in a foundry (Fig. 1)

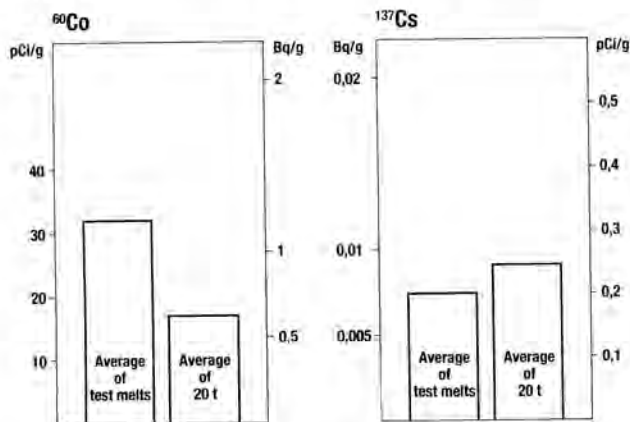


Fig. 1 Comparison Between Radionuclide Concentrations in Test Melt Ingots and in Ingots of Large Scale Melts

The number of test melts depends on the geometry of the decommissioned component. Usually 3 to 6 melt down runs with about 4 kg material each were performed.

Melt experiments with α -contaminated materials

The behavior of α -contaminated scrap originating from a decommissioned suspension test reactor has been tested in the KWU laboratory unit in three melt down runs. From the chemical behavior of actinides one can expect to be oxidized during the melt process and thus released into the slag. Therefore slag additives were used in the experiments. The additives were carbon, siliconcarbide and slag former. Each run contained about 4 kg of scrap. The melt temperature was between 1,480°C and 1,530°C. After cooling down, crucible, ingot and slag were separated. The ingot was divided into measuring specimens as shown in Fig. 2

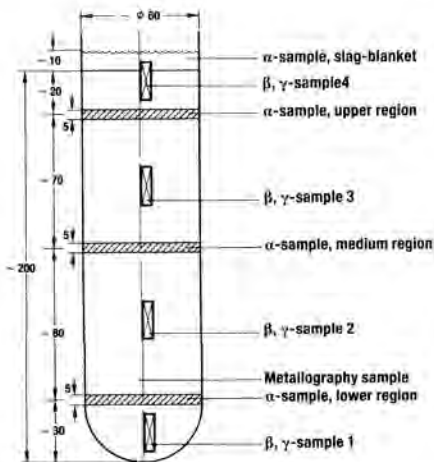


Fig. 2 Ingot of Melted α -Contaminated Scrap Areas for Various Measuring Samples

The only radionuclides which could be measured by γ -spectrometry were Co 60 and Cs 137. The result of this analysis is shown in Fig. 3. Co 60 shows a very homogenous distribution in the test ingots. The broken part of the bars shows the variation in the ingots (refer to Fig. 2). The average of the Co 60 content is marked by an short dashed line with a point. Cs 137 is washed out of the melt into the slag and the off-gas system. The three melts show different residual concentrations which correlate to the measured α -content of the corresponding slags. Obviously the remains in the melt depends on the available total quantity in the contaminated material. The variation in the ingot of melt 3 is large. That comes from one value near the slag zone. This area may contain some inclusions of small slag particles thus pushing up the Cs 137 content drastically.

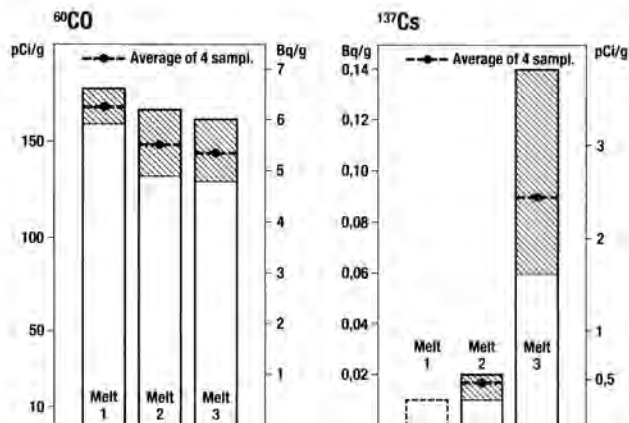


Fig. 3 Co 60 and Cs 137 Content in Test Melt Ingots of α -Contaminated Scrap

Development of a direct α -measuring technique for metallic and slag samples

The intention was to develop a simple economical measuring method in order to control large scale melts regarding their effectiveness on the melt decontamination step. That means that the residual α -activity is well below the limits stipulated by the authorities. Therefore a direct measuring technique for α -radionuclides was developed and tested with the specimens of the three test melts.

As shown in Fig. 2 three specimens (upper, medium and lower area) were cut from each test ingot. These specimens were mounted on a support area and ground and polished with a surface grinding machine. The polished discs had a final diameter of 58 mm and a thickness of 4 mm.

A low level α -measuring device was used for measuring the α -radionuclides in the specimens. The measuring principle is as follows:

background and specimens are measured alternately for 100 minutes during the day. During the night the background or a specimen is measured for 400 minutes. Each specimen is measured for two days; such a measuring procedure is necessary in order to eliminate fluctuations of the background which may reach factors up to 100. This means that each specimen is measured 6 times and the background is measured 8 times.

α -emitters can only be measured in a very thin surface area. Therefore multiple measurements of the specimen are necessary to assure that the activity distribution in the bulk is homogenous. This was obtained by repeatedly grinding off thin layers and subsequent measurement of the α -activity.

According to the origin of the scrap the α -energy of α -radionuclides present in the scrap varies between approximately 4 and 6 MeV. Thus the range of the α -radiation in iron is between approximately 8 and 14 μm (1,2). It was necessary to determine the emission yield in iron and simulated slag samples. So far an Am 241 standard solution has been used for calibration purposes. The simulation of the slag specimen was performed by co-precipitating Am 241 with $\text{Fe}(\text{OH})_3$. The precipitate was afterwards converted to Fe_2O_3 (measuring preparation 1). The simulation of the ingot specimen was performed in two ways. One preparation was performed by suspending iron powder in a few cubic centimeters of an Am 241 standard solution and then evaporating to dryness (measuring preparation 2). Another preparation was performed by impregnation of iron with the Am 241 standard solution and evaporation to dryness (measuring preparation 3). The results of the determination of the emission yield E_e are summarized in table I.

Table I

Determination of the Emission Yield E_e for the α -Emitter Am 241 in Iron and Slag Samples.

Measuring preparation	used activity (Bq/g)	count rate (s^{-1})	corrected count rate ¹⁾ (s^{-1})	range ²⁾ of α -radiation (g/cm^2)	E_e
1	19.15	1.41E-1	1.76E-1	1.119E-2	0.155
2	3.83	4.3 E-2	5.44E-2	9.529E-3	0.285
3	3.83	4.1 E-2	4.94E-2	9.529E-3	0.256

1) Measuring preparations were covered with Hostaphen foil, its influence on the count rate was determined.

2) source: Ref. (2)

The emission yield was calculated according to the following equation:

$$E_e = \frac{Z_n}{C} \cdot \frac{1}{O \cdot E_d \cdot E_g \cdot R} \quad (1)$$

with

- E_e = emission yield
- Z_n = count rate (s^{-1})
- C = activity concentration (Bq/g)
- O = sample area (cm^2)
- E_d = detector efficiency
- E_g = geometric efficiency
- R = range of α -radiation in the sample (g/cm^2)

With the knowledge of the emission yield E_e , the activity concentration C (Bq/g), the standard deviation s and the detection limit g can be calculated according to the following equations

Activity Concentration:

$$C = \frac{Z_n}{O \cdot E_d \cdot E_g \cdot E_e \cdot R} \quad (\text{Bq/g}) \quad (2)$$

Standard Deviation:

$$s = \frac{1}{O \cdot E_d \cdot E_g \cdot E_e \cdot R} \cdot \sqrt{\frac{Z_o}{t_o} + \frac{Z_b}{t_m}} \quad (3)$$

Z_o = count rate background (s^{-1})

Z_b = total count rate (s^{-1})

t_o = measuring time of the background

t_m = measuring time of the sample

Detection Limit:

$$g = \frac{3}{O \cdot E_d \cdot E_g \cdot E_e \cdot R} \cdot \sqrt{\frac{Z_o}{t_o} + \frac{Z_b}{t_m}} \quad (4)$$

The used sample size of 26.4 cm^2 counting area yielded a detection limit of 0.8 Bq/g with a measuring time of 100 minutes. This detection limit can easily be reduced by using larger counting areas.

The α -measurement of the ingot samples (26.4 cm^2 counting area) of the three test melts resulted in agreeing values of < 0.8 Bq/g and < 0.7 Bq/g respectively. That means the α -activity in the melt is below the detection limit.

As can be seen from table II the α -activity is concentrated in the slag samples with some aerosol release to the off-gas system. The aerosols were collected in the off-gas filter during the three test melts.

Table II

Measured α -Activity in Slag Samples and the Off-Gas Filter

test melt	slag quantity g	α -activity concentration Bq/g	total α -activity Bq	
			slag	off-gas filter
1	7.9	1.8	14.2	sum of
2	47.5	3.4	161.5	3 test melts:
3	26.1	3.6	354.5	120

Adding up the activity of the three slag samples and the off-gas filter yields a total α -activity of 650 Bq. This value is in accordance within a factor of 2 compared to the 322 (400) Bq which were measured as α -surface contamination on the material samples used for the test melts. The value in brackets is reached by adding up the detection limit measured at three pieces. Because of the fact that the calibration was only performed with Am 241 which belongs to the group of

α -emitters with rather high α -energies (5.486 and 5.443 MeV) the correlation is much better when the calibration with U 235 (α -energy 4.400 MeV) is considered too. U 235 is one of the major α -radionuclides in the contamination layer.

It can be concluded from the α -measuring results that the real α -activity in the ingots is much lower than the detection limit of < 0.8 Bq/g. Thus the goal to compact and decontaminate α -contaminated scrap in a one step process was reached. The experiments show too that direct α -measuring techniques using large area α -counters can be used to control large scale melt downs in a foundry.

The so far encouraging results will stimulate further experiments. The investigations will be extended to the determination of the emission yield of other α -emitters and to the enlargement of α -counting areas in order to establish a detection limit well below 0.3 Bq/g. A value in such order of magnitude is to be expected to be fixed by the European Community authorities as a limit for the unrestricted re-use of material after melt down of α -contaminated scrap.

Large Scale Melting Processes

KWU in cooperation with Siempelkamp, a commercially operated foundry licenced for melting of low level contaminated metal scrap, developed processes for non-hazardous re-use in order to satisfy the regulation of the German Atomic Energy Act. The activity concentration of the material determines whether or not the parts and components have to be pre-decontaminated by KWU before they are melted in the foundry

The activity concentration is determined on the basis of representative measurements of surface activity and then converted to the mass of the material. In parallel samples are taken from representative parts, melted in the laboratory and the activity subsequently measured (see part 2.1).

If the thus measured and calculated value is well below the maximum volumetric limit stipulated in the license, the entire material is broken up, packed in drums and sent to the foundry for melting.

Proof of its harmlessness is furnished by test measurements made on melted samples in the foundry and on newly cast parts. These test results are verified by the competent authorities.

Replaced fuel racks, moisture separators/reheaters, tube bundles, high-pressure feedheaters and reheater drains coolers with a total weight of 500 t have already been treated for non-hazardous re-use.

The components mentioned above were converted into cast metal drums, shielding elements, crane counterweights etc. for re-use in nuclear facilities.

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