# Nuclide Distribution in the Metal Recycling Process – 14290

Per Lidar*, Maria Lindberg*, Arne Larsson*, and Patrik Konneus*
* Studsvik Nuclear AB
SE-611 82 Nyköping
Sweden
Givenname.familyname@studsvik.se

## ABSTRACT

The Studsvik metal melting facility has been in operation since 1987 for segmentation, decontamination and melting. Thousands of tonnes of metallic low level waste have been processed with the aim of clearance. The nuclide distribution in the ingots of carbon and stainless steel, aluminium, and, lead has been analysed:

Carbon and stainless steel
- 27 700 tonne treated
- Co-60 is the totally dominating nuclide in the ingots (>96%), low energy emitting beta nuclides excluded

Aluminium
- 800 tonne treated
- Co-60 is dominating measured nuclide in the ingots with >60%, low energy emitting beta nuclides excluded

Lead
- 400 tonne treated
- Sr-90 is dominating measured nuclide with >50%, low energy emitting beta nuclides excluded

For 5 200 tonne of the melted carbon and stainless steel, the nuclide distribution in ingots and its secondary waste was investigated in detail with the following confirmations and result:
- The data available was found to be enough to perform a relevant analyze
- It confirms literature data regarding nuclide distribution in the melting process
- Nuclide distribution in ingots, slag, and, dust from decades of melting in Studsvik for selected nuclides was summarized in a table
- Forms an initial platform for development of more precise nuclide transfer models for the melting process.

The main conclusion is that an extensive amount of high quality data exist and that that it is feasible to develop improved models for the nuclide distribution during the melting process based on further analysis of these data and the experience built up.

## INTRODUCTION

Melting of contaminated metallic low level waste from the nuclear industry, as well as other operations involving or generating radioactive isotopes, for release from regulatory control and recycling is an established treatment method.

The nuclide vectors differ between different types of facilities:

- NPP BWR and PWR primary circuit standard operation without significant fuel failures
  - Dominant contaminants activation products are Co-60, Ni-63 and Fe-55
- NPP with fuel failure history
  - Also fission products such as Cs-137, Sr-90, and certain alpha emitting nuclides have to be considered as a significant part of the nuclide inventory
- Fuel factories
  - Uranium isotopes and its progeny
  - TRU-elements and activation products (mainly for MOX plants)
- Research facilities (LWR, HWR, Spallation sources)
  - A wide combination of nuclides can exist

The variation in nuclide composition lead to that specific and reliable knowledge about the nuclide distribution in the material sent for treatment as well as during the metal recycling process is essential for all involved parties up to and including the repository owner.

**Background**

There is an increased focus on the nuclide inventory in the secondary waste resulting from the treatment of metals.

There also seems to be a large conservatism, due to uncertainty, in assigned inventory for metals sent for treatment by melting. This may cause incorrect decisions in the different process steps and costly overestimations of the inventory in the residues sent for disposal, resulting in theoretically filling the repository with radionuclides before it is really full.

Decommissioning projects could be better optimized with

- Less conservative nuclide inventories
- Better knowledge of the transfer of nuclides during melting.

**Purpose**

The purpose of this work was to investigate and understand the real nuclide distribution in the melting process in order to

- Obtain a good characterization of the inventory
- Support the clearance process of the ingots
- Create less conservative nuclide vectors for the secondary waste
- Give better guidelines to decommissioning projects.

**Goal**

The goal is to establish an accepted formula for the nuclide distribution during the melting process.

**METAL RECYCLING PROCESS**

Melting is suitable for low level waste with all types of contaminations. The target is to capture

separated radioactivity either in the slag or in the off-gas treatment system. Nuclides in gas phase at room temperature lead to special requirements for the off-gas treatment.

For the melting, established techniques and facilities are available. Melting of scrap metal has been performed and developed over the centuries and there is a continuous development going on both within the conventional steel industry but also for applications on contaminated metals. For applications on metals contaminated with radionuclides international guidelines and recommendations exists such as EC RP89 [1] regarding conditional clearance of ingots after melting.

The need and interest for melting services has increased significantly over the years. To meet the market as well as regulatory demand several facility upgrades and extensions have been made to the melting facility at Studsvik. Today the licensed capacity is 5 000 tonnes per year. Two induction furnaces are used at the facility; one can be seen in Figure 1.

To allow that the metals, after treatment, can be subject to clearance the regulatory framework and current operating licenses requires validated processes. The extensive knowledge in metal melting and the nuclide distribution in the processes has been an important parameter in this validation process.

Studsvik has all data for the material treated over the decades in a data base, and this paper gives examples of how nuclides will be distributed through the metal recycling process, allowing the possibility to predict the nuclide transfer to the metal ingots as well as to the different fractions of the secondary waste generated in the process.

Fig.1. Induction furnace at Studsvik for melting ingots of contaminated metallic low level waste.

## MATERIAL DIFFERENCES IN NUCLIDE DISTRIBUTION

The nuclide distribution in the melting process is to certain degree material specific. Based on this material specific knowledge the pre-treatment efforts (steel shot blasting etc.) can be optimized.

Generally, the following has been observed, see also Table I:

- Carbon and stainless steel
  - Co, Mn, Fe, Ag and Ni isotopes are closely linked to the steel matrix and stays therefore to a high degree in the metal (the ingot)
  - Heavy elements (U, Am, Pu) are likely to be transferred to the slag (either automatically or by certain special treatment)
  - Substances which evaporates at the actual metal bath temperature are transferred to the slag or the dust (Cs etc.).
- Aluminium
  - Co stays in the metal
  - Most heavy elements (U, Am, Pu) stays in the metal to a high degree
  - Nuclides which evaporates at the actual metal bath temperature are transferred to the slag or the dust.
- Lead
  - Most isotopes, including Co, can be removed from the metal
  - Co end up in the slag
  - Certain elements such as silver (Ag) are difficult to separate in the melting process
  - Heavy elements (U, Am, Pu) ends up in the slag to a high degree
  - Nuclides which evaporates at the actual temperature are transferred to the slag or the dust
  - Special personnel safety arrangements are mandatory when handling lead.

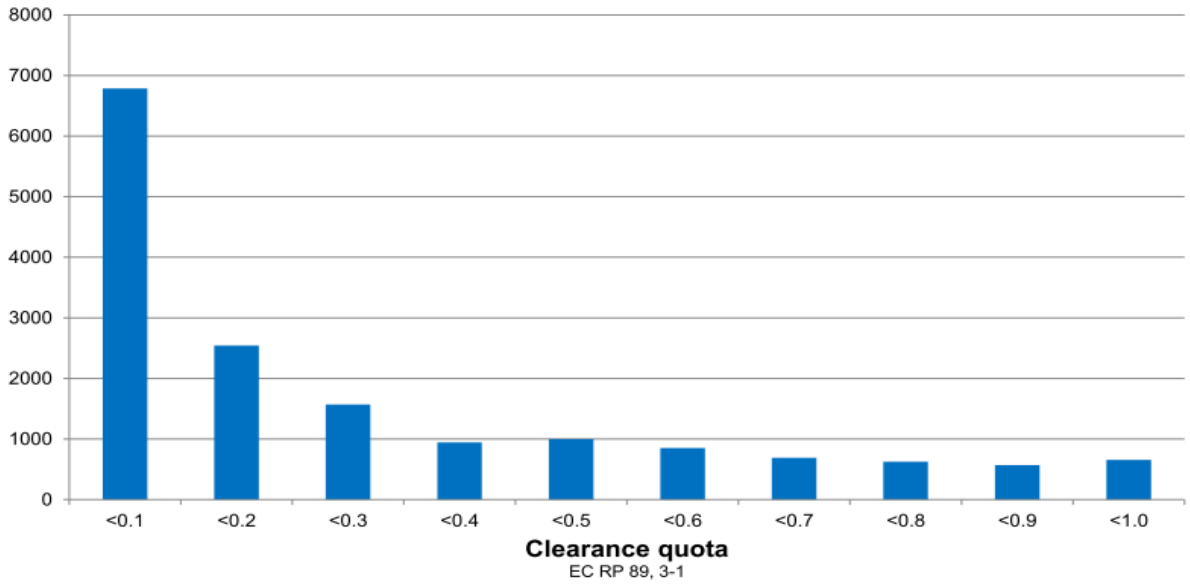


Fig. 2. Clearance quota vs. tonne conditionally cleared ingots produced at Studsvik 2005 – 2012.

Figure 2 shows a histogram over conditionally cleared ingots produced at Studsvik 2005 – 2012, in total approx. 10 000 tonnes are included. The large majority of the ingots have a low clearance quota[i] which indicates a margin to the allowed clearance quota of 1, but it says nothing about the nuclide distribution.

TABLE I. Overview– Nuclide distribution for most important nuclides in steel, excerpt from Table 6.2 in [2] which includes data from different sources.

| Nuclide | Steel (%) | Slag (%) | Dust (%) | Other (%) |
|---|---|---|---|---|
| Mn-54 | 24-100 | 1-75 | 0-5 | 0 |
| Co-60 | 20-100 | 0-1 | 0-80 | 0 |
| Zn-65 | 0-20 | 0-1 | 80-100 | 0 |
| Sr-90 | 0-20 | 95-100 | 0-10 | 0 |
| Ag-108m | 75-100 | 0-1 | 0-25 | 1 (bottom) |
| Sb-125 | 60-100 | 0-20 | 10-40 | 0 |
| Cs-137 | 0 | 0-5 | 95-100 | 0 |
| U | 0-1 | 95-100 | 0-5 | 0 |
| Pu | 0-1 | 95-100 | 0-5 | 0 |
| Am-241 | 0-1 | 95-100 | 0-5 | 0 |

## RESULTS, NUCLIDE DISTRIBUTION IN INGOTS AND SECONDARY WASTE

The results of the nuclide distribution in the ingots melted at Studsvik of carbon and stainless steel, aluminum and lead is shown in Table II. The dominant nuclides from measurements are listed as percentage of total activity. Samples from all ingots are measured with gamma spectrometry, and most of the ingots (depending on the origin) are also analyzed by alpha spectrometry.

The melting results in Table II show the following regarding material differences:

Carbon and stainless steel

- Total amount melted and analysed is 27 700 tonne
- Includes ingots for direct clearance and under decay storage prior to clearance (94% of the total tonnage)
- Nuclide distribution in ingots
  - Co-60 >96% of all activity, low energy emitting beta nuclides excluded
  - Top six nuclides corresponds to 99%

Aluminium

- Total amount melted and analysed is nearly 800 tonne
- Includes ingots for direct clearance and under decay storage prior to clearance (85% of the of the total tonnage)

- Nuclide distribution
  - Co-60 dominating with >60%, low energy emitting beta nuclides excluded
  - Top six nuclides corresponds to 95%

Lead

- Total amount melted and analysed is about 400 tonne
- All ingots were subject for clearance and included in the table
- Nuclide distribution
  - Sr-90 is dominating with >50%, low energy emitting beta nuclides excluded
  - Top six nuclides corresponds to 89%
  - The limited amount of lead treated coming from a limited number of facilities gives an uncertainty in the results.

TABLE II. Measured nuclide content for ingots melted at Studsvik for some material categories.

| **Material category** | **Weight, tonne** | **Measured nuclide content, % of total activity** | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| CS and SS** | 27 749 | Co-60 | Mn-54 | Sb-125 | Zn-65 | Ag-110m | Ru-106 | Uranium | Other alpha | Others |
| | | 96% | 1.1% | 0.7% | 0.5% | 0.3% | 0.3% | 0.2% | 0.1% | 0.4% |
| | | | | | | | | | | |
| Aluminium* | 772 | Co-60 | Zn-65 | Uranium | Mn-54 | Cs-137 | Cr-51 | Na-22 | Sb-125 | Others |
| | | 62% | 11% | 6.4% | 5.6% | 4.6% | 4.2% | 1.4% | 1.1% | 2.8% |
| | | | | | | | | | | |
| Lead* | 395 | Sr-90 | Ag-110m | Cs-137 | Co-60 | Ag-108m | Am-241 | Pu-239 | Sb-125 | Others |
| | | 52% | 17% | 5.8% | 5.5% | 5.0% | 3.9% | 3.1% | 1.3% | 6.3% |

*) Minimum detectable activity ((MDA) values are included from gamma / alpha spectrometry.

**) Carbon Steel (CS); Stainless Steel (SS).

For carbon and stainless steel, since Co-60 and Cs-137 are the dominating gamma emitting nuclides as well as correlation nuclides for hard to measure nuclides, it was decided to initially concentrate further analyses on these two nuclides. 5 200 tonne material from different LWRs were analysed with regards to the relation of activity in pre-treatment residues / ingots / slag / dust. The result is shown in Table III. Containerized scrap, i.e. scrap from maintenance or decommissioning activities, and large components are separated. The minimum, maximum and median values of the measured activity fraction in the ingots, dust and slag are listed.

As can be seen in Table III, there seems to be a difference between the nuclide distributions of Co-60 and Cs-137 in steel recycling from containerized scrap respectively large components. The difference could be a result of several factors, such as different surface to volume relationship between incoming waste in the groups, and differences in the effectiveness of pre-melting decontamination.

It should be noted that the measured activity is based on gamma and alpha spectrometry. Hard to measure nuclides (such as low energy beta emitters) are not measured in the metal, but is rarely any issue for the conditional clearance of ingots since the clearance values are so high compared to the ones for Co-60. However, the distribution of the pure beta emitters has to be known and considered, since the long lived nuclides can be important for the disposal long term safety case.

TABLE III. Nuclide distribution of Co-60 and Cs-137 in carbon and stainless steel recycling at Studsvik.

| **Co-60** | | | | | **Cs-137** | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Container scrap | | Max* | Min* | Median** | Container scrap | | Max* | Min* | Median** |
| Blasting residues | | 91% | 18% | 69% | Blasting residues | | 16% | 1% | 14% |
| Distribution of the remaining activity in the melting process: | | | | | Distribution of the remaining activity in the melting process: | | | | |
| Melting fraction | Ingots | 92% | 29% | 67% | Melting fraction | Ingots | 0% | 0% | 0% |
| | Dust | 58% | 6% | 24% | | Dust | 70% | 23% | 57% |
| | Slag | 14% | 2% | 4% | | Slag | 77% | 30% | 52% |
| Large components | | Max | Min | Median | Large components | | Max | Min | Median |
| Blasting residues | | 82% | 14% | 46% | Blasting residues | | 35% | 10% | 20% |
| Distribution of the remaining activity in the melting process: | | | | | Distribution of the remaining activity in the melting process: | | | | |
| Melting fraction | Ingots | 99% | 88% | 97% | Melting fraction | Ingots | 0% | 0% | 0% |
| | Dust | 7% | 0% | 2% | | Dust | 29% | 10% | 15% |
| | Slag | 6% | 1% | 2% | | Slag | 90% | 71% | 85% |

*) The max and min values are average values for each delivery.
**) The sum of median values may not end of to 100% due to limited number of deliveries compared.

## LITERATURE COMPARISON

The literature comparison is primarily made against two references [2, 3], see Table IV. The first reference [2] is applicable to steel. In Table 6.2 information is combined from different sources (Cheng et al., 2000; Nieves et al., 1995; NRC, 1999), leading to large uncertainty for some nuclides. Different furnace types (basic oxygen furnace and electrical arc furnace) are included in the compilation.

The second reference [3] is reflecting the experience from a facility in Germany and is applicable to steel. Detailed nuclide distribution for many nuclides is provided.

The median values taken from Table III are re-scaled in Table IV to achieve a total sum of 100%.

TABLE IV. Comparison of literature and Studsvik data of nuclide distribution during metal recycling process (carbon and stainless steel).

| Nuclide | Ref. | Note | Steel (%) | Slag(%) | Dust (%) | Other (%) |
|---|---|---|---|---|---|---|
| Co-60 | [2] | | 20-100 | 0-1 | 0-80 | 0 |
| | [3] | | 88 | 11 | 1 | 0 |
| | Container | Min - Max | 29-92 | 2-14 | 6-58 | 0 |
| | Container | Median* | 70 | 4 | 26 | 0 |
| | LC | Min – Max | 88-99 | 1-6 | 0-7 | 0 |
| | LC | Median* | 96 | 2 | 2 | 0 |
| Cs-137 | [2] | | 0 | 0-5 | 95-100 | 0 |
| | [3] | | <1 | 60 | 40 | 0 |
| | Container | Min - Max | 0-<1 | 30-77 | 23-70 | 0 |
| | Container | Median* | 0 | 47 | 53 | 0 |
| | LC | Min - Max | 0-<1 | 71-90 | 10-29 | 0 |
| | LC | Median* | 0 | 85 | 15 | 0 |

*) Median value re-scaled to 100% total sum.

## CONCLUSION

Studsvik has for more than three decades treated and melted mainly carbon and stainless steel but also aluminium, copper, brass and lead. Over the years has an extensive experience been built and data been collected.

Based upon an analysis of the extensive dataset it has been shown that it is feasible to develop improved models for the nuclide distribution during the melting process based on analysis of these data and the experience built up. This is important in order to support both waste generators and disposal organisations in their task to optimise their processes without violating the short or long term safety.

## REFERENCES

1. Radiation protection 89, recommended radiological protection criteria for the recycling of metals from the dismantling of nuclear installations, European Commission, 1998.

2. NCRP (2002). Managing Potentially Radioactive Scrap Metal, NCRP Report No 141, NCRP, Bethesda, MD.

3. QUADE, U. and MÜLLER, W. (2005). Recycling of radioactively contaminated scrap from the nuclear cycle and spin-off for other application, Rev. Metal. Madrid Vol. Extr. (2005) 23-28.

[1] In a practical case with more than one radionuclide involved, the clearance quota is defined as follows (Ref.: European Commission Radiation Protection 89):

To determine if a mixture of radionuclides is below the clearance level a summation formula is used:

$$\sum_{i=1}^{n} \frac{c_i}{c_{Li}} < 1.0$$

where

$c_i$ is the specific activity of radionuclide *i* in the material being considered (Bq/g and Bq/cm2),
$c_{Li}$ is the specific clearance level of radionuclide *i* in the material (Bq/g and Bq/cm2),
$n$ is the number of radionuclides in the mixture.

In the above expression, the ratio of the concentration of each radionuclide to the clearance level is summed over all radionuclides in the mixture. If the sum (clearance quota) is less than one the material complies with the clearance requirements.