Residual Materials Containing Radioactivity Detected in Spanish Scrap Yards and Melting Installations. The ENRESA Experience

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Abstract. Throughout the last six years several materials with radioactive content have been detected in Spanish metal scrap yards as well as steel melting installations. The detection of these materials is the result of a global programme called “Spanish Protocol for Collaboration on Radiation Monitoring of Metallic Materials”. The object of this programme is to perform radiological controls on initial metallic materials and final products in order to detect the presence of radioactive materials. This programme has been widely implemented in Spain after the incident with a Cs-137 source in a melting facility. The Protocol is complemented by a Transfer Authorisation published by the Ministry of Economy (February 2000) which establishes radioactivity levels for classifying the detected radioactive materials as radioactive waste. The task of the Spanish National Company for Radioactive Waste Management (ENRESA) is to recover and dispose in suitable installations the detected materials classified as radioactive waste. In this paper are described the activities carried out by ENRESA during the last six years, together with a description of the materials involved, their radiological properties (isotopic composition and activity content, dose rate), physical characteristics, and their final management.

1. Introduction

After the incident with a Cs-137 source in a Spanish melting facility in June 1998 the use of portal monitors has been extended in metal scrap yards and steel melting installations. The task of these detectors is to prevent that radioactive sources and materials with radioactive content enter the facility and avoid that their processing. In general, the detectors are placed at the entrance of the installations and alarm when the radioactive level gets higher than the background level.

When there is an alarm, first it is necessary to perform an investigation to determine whether it is false or not. If the existence of radioactive material is confirmed, it is located, segregated and put in conditions of safety. After that, the best way to manage the material should be found, either by returning to the supplier, or transferring it to the Spanish Radioactive Waste Management Company (ENRESA).

With the object of establishing a coherent control system and clarifying the steps to undertake in case of an alarm, a global programme was prepared and implemented: “Spanish Protocol for Collaboration on Radiation Monitoring of Metallic Materials” (Protocol) [1].

2. Protocol for Collaboration on Radiation Monitoring of Metallic Materials

The object of this protocol is to establish requirements for radiation monitoring of metal materials and final products. These requirements aim at detecting radioactive materials and avoiding the risk that they should be dispersed, and consequently, people, property and the environment in general be exposed to radiation or contamination.

The Protocol was signed in November 1999 by all the parties concerned: the corresponding Ministries, the Spanish Regulatory Body (Nuclear Safety Council, CSN), the Spanish National Company for Radioactive Waste Management (ENRESA), the Trade Unions and companies that recover, handle, store and recycle metallic scrap. The Protocol includes a technical annex where the conditions for the radiological control of the initial metallic materials, final products and by-products (slags and off-gas dust) are established, together with the activities that should be carried out when radioactive materials are detected.

All the signatory companies are included in a Register set up by the Ministry concerned. These companies must provide a declaration containing the following information:
- Name of signatory company.
- Description of the facilities.
- Description of monitoring and control system.
- Person responsible for radiation monitoring in the facility.
- Declaration of acceptance of the Protocol.

The companies included in this programme have the following responsibilities:

- To install a monitoring system to detect any kind of radioactivity in the scraps before entering the plant.
- To demand a Non-Radioactivity Certificate before accepting a cargo from foreign countries.
- To control the final products and by-products with suitable equipments.
- To inform CSN about all the alarms related to radioactive materials.
- To sign a contract with ENRESA for removal of detected radioactive materials.

The Protocol also includes investigation levels, which specify what activities must be carried out in case they are exceeded, as well as exemption levels. The radioactive materials exceeding the exemption levels must be transferred to ENRESA for their management as radioactive waste.

The type of activities to be carried out depends on where the radioactive material has been detected. When the radioactive material is in a cargo entering the installations the actions are the following:

- Isolating the cargo.
- Monitoring the contents of the cargo to detect the radioactive material.
- Segregation and storage of the detected radioactive material in safe conditions.
- Notifying the CSN.
- Taking care of the radioactive materials until they are removed by ENRESA.

Should the radioactive materials be detected in the final products or by-products, it is necessary to analyse them. If the activity levels are found to be above the established levels, then it is necessary to stop the production and proceed to assess the situation, previously notifying the CSN. Later the company in question must prepare a programme for recovering the facility and conditioning the produced radioactive wastes before they are removed by ENRESA.

The personnel of the factory can carry out all these activities either alone or with the help of a Radiation Protection Unit (RPU), which is authorised by the CSN and is responsible for fulfilment of the Radiation Protection Regulations. The ENRESA RPU carries out all necessary activities for removal of materials considered to be radioactive wastes.

The Protocol is complemented by a Transfer Authorisation published by the Ministry of Industry and Energy in February 2000 [2], which contains the investigation levels and the exemption criteria.

The investigation levels are applicable to the monitoring systems. They are the following:
- For automatic portal monitors: three standard deviations from the background radiation.

- For manual methods of measurement: more than 0.3 µSv/h in contact with the vehicle surface.

- For final products or by-products: the levels applicable are a fraction of the exemption levels.

According to the exemption criteria the materials that did not have to be submitted to regulatory control or which proceed from an authorised release are considered exempted. Also, the Transfer Authorisation contains specific radioactivity levels proposed by the CSN for classifying the detected radioactive materials as radioactive waste. These values are based on the recommendations of the European Commission for recycling of metals resulting from dismantling of nuclear installations (“Radiation Protection nº 89”). The materials classified as radioactive waste are collected and disposed in suitable installations by ENRESA, while the materials with low levels are processed at the very same facility.

In this Authorisation, there are some problems still to be solved: for example, the issue of the low activity sources and the naturally occurring radioactive materials (NORM). These materials can be processed at the installations although their concentration levels are above the value established in the Transfer Authorisation (1 Bq/g) because of the quantities involved are very small. At this moment, the materials contaminated by NORM above the reference level are temporarily stored at the facilities for future melting in case the CSN approves the new levels, which ENRESA proposed in a specific study.

3. ENRESA activities

In the last six years the ENRESA RPU carried out in 102 actions in 33 steel melting facilities and metal scrap yards. These actions can be classified in two types: immediate and planned. The immediate actions are the ones carried out with some urgency and are mainly oriented to location, segregation, isolation and even prompt removal of the detected radioactive material; or to establishing the radiological conditions at the installation after an incident. In some cases these kinds of actions are carried out at the requirement of the CSN. The planned ones are carried out after the transfer of the radioactive material to ENRESA and are oriented to identifying, characterizing and conditioning the materials before removal. In general, several facilities in the same geographical area are visited together. Apart from these actions ENRESA has carried out 75 collections and transportations of the detected radioactive materials in 33 facilities. The yearly distribution of the actions carried out is shown in Table I.

<table>
<thead>
<tr>
<th>Year</th>
<th>Actions</th>
<th>Collections</th>
</tr>
</thead>
<tbody>
<tr>
<td>1998</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>1999</td>
<td>18</td>
<td>11</td>
</tr>
<tr>
<td>2000</td>
<td>14</td>
<td>15</td>
</tr>
<tr>
<td>2001</td>
<td>10</td>
<td>8</td>
</tr>
<tr>
<td>2002</td>
<td>24</td>
<td>14</td>
</tr>
<tr>
<td>2003</td>
<td>33</td>
<td>26</td>
</tr>
</tbody>
</table>

In this period 11 immediate actions have been carried out: two of them were related to the incidental melting of Cs-137 sources in two melting facilities and one to an incident in a scrap yard, also with a Cs-137 source. The other 8 consisted in segregation and removal of medium activity radioactive sources. In three cases the sources did not have a shielding.

The activities carried out in case of an incident are radiological surveillance of the plant (including sample-taking) in order to assess the radiological risk; and initial assess of the radioactive waste volume, its activity and isotopic composition. In general, in the immediate interventions (not
incidents) the activities start with localisation of the material in the scrap; afterwards, the radioactive material is segregated and conditioned for transport. If the activity of the source is relevant or other circumstances advise it, the radioactive material is removed as soon as possible.

The planned actions cover all the activities needed to characterize the radioactive material and define whether it must be removed as a radioactive waste. Gamma spectrometry and different kind of measurements are applied in order to determine the activity of the source.

4. Characteristics of detected radioactive materials

During these years more than 500 pieces and about 86 sources of different activity have been reviewed. Most of them were contaminated by NORM and could be melted. In Figure 1, the distribution of the reviewed materials is presented.

![Distribution of detected materials](image1.png)

The main types of this kind of material are the following:

- Radioactive sources: in general (about 45%), the activity of the sources is low (below 1Bq/g) and the most common radionuclide is Ra-226 (71%). The most frequently detected relevant sources are Cs-137 or Co-60 sources. Other sources have also been detected: Am-241, Am/Be, Ra/Be and Kr-85. Figure 2 shows the distribution of detected sources and figure 3 their activity distribution.

![Distribution of detected sources](image2.png)
The radiological characteristics of the most relevant sources are included in table II.

Table II Characteristics of relevant detected sources

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Number</th>
<th>Activity (MBq)</th>
<th>Dose rate (mSv/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Minimum</td>
<td>Maximum</td>
</tr>
<tr>
<td>Am-241</td>
<td>3</td>
<td>4.4 E2</td>
<td>3.7 E3</td>
</tr>
<tr>
<td>Am-241/Be</td>
<td>1</td>
<td>-</td>
<td>2.6 E3</td>
</tr>
<tr>
<td>Co-60</td>
<td>6</td>
<td>4.3 E1</td>
<td>1.1 E3</td>
</tr>
<tr>
<td>Cs-137</td>
<td>11</td>
<td>6.5 E1</td>
<td>6.26 E3</td>
</tr>
<tr>
<td>Kr-85</td>
<td>1</td>
<td>-</td>
<td>1.85 E3</td>
</tr>
<tr>
<td>Ra-226</td>
<td>10</td>
<td>3.9 E0</td>
<td>5.1 E2</td>
</tr>
<tr>
<td>Ra-226/Be</td>
<td>1</td>
<td>-</td>
<td>1.1 E2</td>
</tr>
</tbody>
</table>

(1) Sources shielded

- Consumer goods: the most common consumer goods detected are devices with luminous dials (containing radium), lightening rods (containing radium or americium), smoke detectors, electronic components (containing radium), thorium-magnesium alloys etc. The radiological characteristics of these materials are shown in table III.

Table III Characteristics of consumer goods

<table>
<thead>
<tr>
<th>Type</th>
<th>Radionuclide</th>
<th>Number</th>
<th>Activity (MBq)</th>
<th>Dose rate (µSv/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lightening rods</td>
<td>Ra-226/Ra-226/Am-241</td>
<td>16</td>
<td>1 37</td>
<td>100</td>
</tr>
<tr>
<td>Smoke detectors</td>
<td>Ra-226</td>
<td>21</td>
<td>0.1 2</td>
<td>10</td>
</tr>
<tr>
<td>Luminous dials</td>
<td>Ra-226</td>
<td>51</td>
<td>0.1 8</td>
<td>5</td>
</tr>
<tr>
<td>Alloys</td>
<td>Th-232</td>
<td>9</td>
<td>-</td>
<td>5</td>
</tr>
<tr>
<td>Other goods</td>
<td>Ra-226/Th-232</td>
<td>58</td>
<td>0.01 0.2</td>
<td>0.4</td>
</tr>
</tbody>
</table>

- Depleted uranium used in shieldings and counterweights. These materials presented a contact dose rate between 10 and 50 µSv/h.

- Metallic scraps contaminated with radioactive materials: most of the pieces are contaminated by NORM (figure 4), but sometimes pieces are contaminated by Cs-137 or containing Co-60 (perhaps from an accidental melting in the past). The activity concentration of contaminating material (NORM) is, in general, between 0.1 and 50 Bq/g, although in some cases higher activities have been measured. These values produce an activity concentration on the pieces between 0.1 Bq/g and less than 100 Bq/g,
in the worse case. The general radiological characteristics of the pieces contaminated by NORM are in Table IV.

<table>
<thead>
<tr>
<th>Type</th>
<th>Number</th>
<th>Dimensions (cm)</th>
<th>Dose rate (μSv/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pipes</td>
<td>93</td>
<td>Thickness/Windiness</td>
<td>Length</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.2-1</td>
<td>&lt;100-200</td>
</tr>
<tr>
<td>Pieces</td>
<td>429</td>
<td>0.2-2</td>
<td>100-200</td>
</tr>
</tbody>
</table>

Fig. 4. Metallic scraps contaminated by NORM

- Pieces containing natural radioactive materials as bricks, fireproof materials, concrete with high contents in radium, etc. and residual materials (sands, soils, etc).

All the radioactive sources and materials containing radium and thorium different from NORM were collected by ENRESA and stored in the Spanish Radioactive Wastes Disposal Facility (C.A. Cabril). Only one source was returned to the supplier directly from the melting facility.

5. Conclusions

- The control of the radioactive materials in metallic scraps requires a well-consolidated programme, which defines the responsibilities of all the parts involved, together with the activities to be carried out. The “Spanish Protocol for Collaboration on the Radiation Monitoring of Metallic Materials” and the “Transfer Authorisation” meet these objectives.

- The ENRESA RPU can take part in immediate and planned actions and as usual to identify and characterise the radioactive materials before their removal as radioactive wastes.

- The number of actions carried out annually is not high although a small increase is being observed. In general the detected materials are contaminated by NORM.

- The tasks of identifying, segregating and characterising the radioactive materials required a high professional level and considerable experience.
- It is necessary to establish a new set of exemption values for materials contaminated by NORM.

6. References


2. Resolution to transfer to ENRESA radioactive materials detected in scraps or during the melting process. Spanish Ministry of Industry and Energy. (February 2000).