RISK ASSESSMENT FOR CHEMICAL PICKLING OF METALS CONTAMINATED BY RADIOACTIVE MATERIALS
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In recent years, many cases of contamination of metal scraps by unwanted radioactive materials have occurred. Moreover, international organisations are evaluating the possibility to re-use or to recycle metals coming from nuclear power plants. The metal recycling industry has started to worry about radiation exposure of workers that could be in contact with contaminated metals during each manufacturing phase. Risks are strongly dependent on the radiation source features. The aim of this study is to perform risk assessment for workers involved in chemical pickling of steel coils. Monte Carlo simulations have been performed, using the MCNP package and considering coils contaminated with ⁶⁰Co, ¹³⁷Cs, ²⁴¹Am and ²²⁶Ra. Under the most conservative conditions (coil contaminated with 1.0 kBq g⁻¹ of ⁶⁰Co), the dose assessment results lower than the European dose limit for the population (1 mSv y⁻¹), considering a maximum number of 10 contaminated coils handled per year. The only exception concerns the case of ²⁴¹Am, for which internal contamination could be non-negligible and should be verified in the specific cases. In every case, radiation exposure risk for people standing at 50 m from the coil is widely <1 mSv y⁻¹.

INTRODUCTION
Radioactive materials may contaminate industrial metallic products(¹,²). Moreover, international organisations are evaluating the possibility to re-use or to recycle metals coming from nuclear power plants(³–⁸).

Both situations require to perform radiation risk assessment for workers who could be in contact with the contaminated metal during each manufacturing phase.

In this work, the external and internal radiation risk for workers who perform metal chemical pickling operations (chemical pickling of steel foils rolled up in coils) has been evaluated.

MATERIALS AND METHODS
Two different scenarios should be considered in the radiation risk assessment:
• external radiation exposure from metallic objects;
• internal contamination with radioactive materials released by the metal.

In both cases the risk assessment is strongly dependent on the source characteristics, in other words risk is correlated with the radionuclide that has contaminated the metal and with its specific activity (concentration). There is a large number of radionuclides that could be potentially involved, owing to the wide use of radioactive materials in the modern society.

In many radioactive contamination accidents that take place during industrial melting process the resulting material is uniformly contaminated with non-removable contamination. During manufacturing operations, workers may be affected by external radiation exposure, and also by internal contamination when particular working tasks are performed(⁹–¹⁰). In Table 1 the most common radionuclides that contaminate metallic products in the United States and in Canada are shown. Typical activities for melted radioactive sources range from

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Number of findings</th>
</tr>
</thead>
<tbody>
<tr>
<td>¹³⁷Cs</td>
<td>52</td>
</tr>
<tr>
<td>Th</td>
<td>23</td>
</tr>
<tr>
<td>U</td>
<td>17</td>
</tr>
<tr>
<td>²⁴¹Am</td>
<td>14</td>
</tr>
<tr>
<td>⁶⁰Co</td>
<td>6</td>
</tr>
<tr>
<td>⁸⁵Kr</td>
<td>3</td>
</tr>
<tr>
<td>⁹⁰Sr</td>
<td>2</td>
</tr>
<tr>
<td>³⁵H</td>
<td>2</td>
</tr>
<tr>
<td>Radium</td>
<td>116</td>
</tr>
<tr>
<td>Other unidentified sources</td>
<td>9</td>
</tr>
<tr>
<td>Total</td>
<td>244</td>
</tr>
</tbody>
</table>

Other radionuclides different from those listed in the table: ²¹⁰Pb, depleted Uranium, ²³²Th.

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fraction of GBq to some TBq (9), while the activities of the final products, coming from the melting process, are some orders of magnitude lower (1). In Table 2 some radionuclide concentrations and doses, measured in contact with contaminated metals, are shown.

Manufacturing operations correlated to chemical pickling

Metal scrap is initially collected from several suppliers, processed through a variety of methods (sorting, shredding, shearing, bailing and briquetting) and controlled before being sent to the melting plant. Scrap is melted into new metallic products, typically steel foils. Steel foils are rolled up in coils to be easily transported from the melting plant to the plant where they will be processed.

Before starting a new manufacturing operation, the metal needs to be descaled. Chemical pickling is a procedure of cleaning up the surface of metals: first the foils are unrolled, then they pass through a HCl dip and finally they are rolled in coils again.

In general, metallic products that are the objects of chemical pickling are steel foils, having density of 7.85 g cm$^{-3}$; during chemical pickling process, foils lose ~0.6% of their weight.

Chemical pickling of contaminated metallic products is worth being assessed for radiation risk, since it may involve both risks of external radiation exposure and internal contamination.

To evaluate the dose, a complete analysis of the manufacturing cycle and a full understanding of the activities performed by the workers involved in the coils manipulation have been carried out. Worker activities have been divided into single tasks and for each of them a large number of parameters have been evaluated, such as the number of workers involved, the average distances from the contaminated item, the duration of the operation and the personal protective equipment used.

The manufacturing operations performed in chemical pickling can be divided into the following:

- unloading from transport;
- transporting of the item to a temporary storage area;
- finding and recovering of the item from the storage area;
- preparation of the item for chemical pickling line (worker responsible for the entering and exiting control points of the process line);
- chemical pickling: foil is automatically unrolled, immersed in HCl dip and, at the end, rolled up again;
- preparation for possible coil cut and packaging;
- loading onto transport;
- HCl re-treating and waste processing.

Some activities take place without the direct worker manipulation, others require that the worker manipulates the item standing close to it.

Monte Carlo simulations

Coil dimensions are comparable with the size of a man who is working close to it. Auto-shielding
effect has to be taken into account because of coil thickness. Monte Carlo simulations have been performed, using the MCNP package (version 4c)\(^{(11)}\), in order to evaluate ionising radiation exposure due to contaminated metals with unusual dimensions, such as the coils of the chemical pickling process.

In Monte Carlo simulations the following coil features have been assumed:

- **material**: steel AISI 1040, composed of C 0.4%, Fe 98.6%, Mn 0.07%, P 0.01% and S 0.02%, with density 7.85 g cm\(^{-3}\);
- **geometry**: hollow cylinder with external diameter 1.9 m, internal diameter 0.76 m and height 1.4 m;
- **cylinder position**: suspended in air or laid on a 0.5 m thick concrete floor.

In this evaluation the following radionuclides (taken individually) have been considered, since they represent several spectral emissions of the isotopes as listed in Table 2: \(^{60}\)Co, \(^{137}\)Cs (in equilibrium with \(^{137}\)mBa), \(^{241}\)Am and \(^{226}\)Ra (in equilibrium with \(^{226}\)Rn, \(^{218}\)Po, \(^{214}\)Pb, \(^{214}\)Bi and \(^{214}\)Po). We point out that \(^{226}\)Rn can be considered in equilibrium with \(^{226}\)Ra; indeed, the period of time elapsed between the melting of scrap metal and the chemical pickling is at least 2 weeks (the time necessary to transport the coils from the melting plant to the plant where they will be processed).

The specific activity considered for each of the four radionuclides is 1.0 kBq g\(^{-1}\), uniformly distributed, which corresponds to a total coil activity of 26 GBq. Evaluated radiation exposures are directly proportional to the activity in the manufacturing item. Therefore, the results can be easily scaled for concentrations different from the assumed one.

The physical parameters of radioactive decay (emissions, type of radiation, energy, half-life, etc.) have been taken from several sources\(^{(12-15)}\).

In the simulation, photon fluence, \(\Phi\), and ambient dose equivalent, \(H'(10)\), have been calculated in different positions: in the middle of the cylinder and at some distances from its external surface (on the external surface, at 0.5, 1.0, 5.0, 10.0 and 20.0 m) along the cylinder axis and along a horizontal line crossing the center of the cylinder and orthogonal to the axis itself.

In the case of \(^{60}\)Co and \(^{137}\)Cs isotopes, the latter having a relative high \(\beta\) emission, dedicated simulations with a \(\beta\) source have been performed to evaluate the contribution of Bremsstrahlung from \(\beta\) decay to the total photon fluence, at 1.0 m from coil surface in radial direction. For \(^{60}\)Co the main primary electron emitted in the decay has been considered. For \(^{137}\)Cs the most frequent \(\beta\) of internal conversion (yield 7.77%, energy 624.2 keV) has been generated, as well as the two main primary electrons emitted in the decay. To be sure of not underestimating the effect, an energy equal to the end-point of the \(\beta\) distribution has been attributed to the \(\beta\) source.

MCNP calculates the photon fluence as the average number of photons that hit a infinitesimal spherical surface placed at a point of the space, for emitted photon. Once this quantity is normalised to each radionuclide disintegration and distributed over the energy of the incident photons, it returns the spectral fluence at the chosen point. From the spectral fluence per unit of activity, the ambient dose equivalent at a depth of 10 mm in the human tissue, \(H'(10)\), has been calculated using ICRP\(^{(16)}\) conversion coefficients. The calculated \(H'(10)\) represents the effective dose, assuming that the external exposure is uniform at least in first approximation. Further details about calculations performed in the simulations are reported in Appendix A.

### RESULTS AND DISCUSSION

The calculated spectral fluences due to the four radionuclides considered are shown in Figures 1–4, at a point placed 1.0 m from the surface, in radial direction. The coil is suspended in air. The relative intensity of each peak is, with a good approximation, proportional to the emission probabilities, at least for high energies (>1 MeV). In the low energy region, photoelectric absorption in steel becomes important and reduces the peak intensities.

The spectral fluence due to the Bremsstrahlung contribution for \(^{137}\)Cs is maximum in the energy range around 100–200 keV. Here it is about a factor 20 less than the value calculated for a pure gamma emission; moreover, the integrated ambient dose equivalent is a factor 10\(^3\) less than the value calculated for a pure gamma emission. This result is correlated to the absorption effects due to the considerable thickness of the coil, that is supposed to
be uniformly contaminated. Therefore, this contribution has not been considered for the final dose evaluation. In the case of $^{60}$Co the Bremsstrahlung contribution is much lower than in the case of $^{137}$Cs. In Figure 5 the calculated ambient dose equivalent rates $H^*_E(10)$ (in mSv h$^{-1}$), owing to the four radionuclides considered, are shown for different radial distances from the coil surface, when it is suspended in air. As expected, for the same activity level, $^{60}$Co gives the highest radiation exposure.

In Table 3 ambient dose equivalent rates $H^*_E(10)$ calculated at different points (on the external surface and at different distances both in radial and axial directions), assuming the coil contaminated by $^{60}$Co and laid on the concrete floor, are shown. These values have been used to calculate the effective doses received by the workers who perform their activities close to the coil.

In Table 4, values of $H^*_E(10)$ for the four radionuclides at 0.5 m from the coil surface are shown, both for radial and axial directions. To allow for a comparison, in the same table, the ratios between irradiations due to the considered radionuclides and those due to $^{60}$Co are shown. The results obtained using the described method are in a good agreement with the values found for scenarios correlated to the recycling of contaminated metals coming from the nuclear fuel cycle$^{(5)}$.

To summarise the results of the simulation, it is possible to state the following:

- the highest radiation exposure close to the coil is due to $^{60}$Co contamination, as expected;
- the irradiation percentages relative to $^{60}$Co are 21, 55 and 0.1%, respectively, for $^{137}$Cs, $^{226}$Ra and $^{241}$Am (Table 4);
- no substantial difference has been found in the radiation exposure between the case in which the coil is suspended in air and the case in which it lays on a concrete floor (Table 4);
- the radiation exposure is negligible for distances $>20$ m from the coil surface.

Figure 2. $^{137}$Cs, steel coil suspended in air. Photon fluence per energy interval ($dE = 10$ keV), at 1.0 m from the cylinder surface in the radial direction.

Figure 3. $^{241}$Am, steel coil suspended in air. Photon fluence per energy interval ($dE = 1$ keV), at 1.0 m from the cylinder surface in the radial direction.

Figure 4. $^{226}$Ra, steel coil suspended in air. Photon fluence per energy interval ($dE = 10$ keV), at 1.0 m from the cylinder surface in the radial direction.

Figure 5. Steel coil suspended in air. Ambient dose equivalent rate, $dH^*_E(10)/dt$, vs. radial distance from coil surface, for the four considered radionuclides.
Evaluation of the dose due to external radiation

Evaluations presented in this work are strongly dependent on the assumed hypotheses and, in particular, on the radionuclide and its concentration, the manufacturing cycle, the work task and its duration, the distance from the machined coil and the total number of contaminated coils per year. The external radiation exposure of the workers have been evaluated considering the most conservative condition: the item is assumed contaminated with $1.0 \text{ kBq g}^{-1}^{60}\text{Co}$.

The calculation of the potentially absorbed dose has been implemented considering a worker who stands close to the item (contaminated coil) for a certain period of time and in a position defined in the work task specification. Taking into account that the worker position is extremely variable, the average dose between the radial and the axial position has been adopted (Table 3).

The effective dose $E$, in mSv, received in a shift by the worker, involved in any work task, is composed of two contributions. The first contribution is related to the carrying out of all the activities of a specific task, whereas the second one is due to the presence of items, waiting for the processing, that may be stored close to the worker. The following formula for the effective dose calculation has been used:

$$E = N \sum_n T_n H_n + T_{\text{rem}} H_{\text{rem}}.$$  \hspace{1cm} (1)

where $\sum_n T_n + T_{\text{rem}}$ is the total shift duration (8 h) and:

- $N$ = number of items that are assumed being contaminated and that are handled by the worker in each shift (e.g. $N = 1$ when the calculation is performed considering one item).
- $T_n$ = period of time (h) needed to carry out the $n$th activity of the considered work task;
- $H_n$ = ambient dose equivalent rate $H^*(10)$ (mSv h$^{-1}$) calculated in the position in which the $n$th activity is carried out;
- $T_{\text{rem}}$ = duration (h) in a shift of the activity, called 'everything else in the shift', in which there is not a direct contact with the item;
- $H_{\text{rem}}$ = ambient dose equivalent rate $H^*(10)$ (mSv h$^{-1}$) calculated in the position in which the activity called 'everything else in the shift' is carried out.

It is worth noticing that the effective dose $E$ received in a shift by the worker is not simply

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### Table 3. Ambient dose equivalent rate at different distances from the surface of a steel coil contaminated by $1.0 \text{ kBq g}^{-1}^{60}\text{Co}$ source laying on a concrete floor.

<table>
<thead>
<tr>
<th>Direction</th>
<th>Ambient dose equivalent rate $H^*(10)$, mSv h$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0 m</td>
</tr>
<tr>
<td>Radial</td>
<td>0.675</td>
</tr>
<tr>
<td>Axial</td>
<td>0.755</td>
</tr>
<tr>
<td>Average</td>
<td>0.72</td>
</tr>
</tbody>
</table>

### Table 4. Ambient dose equivalent rate from a steel coil contaminated by different radionuclides (separately) at a distance of 0.5 m.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>$H^*(10)$ at 0.5 m mSv h$^{-1}$</th>
<th>Coil position</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{60}\text{Co}$</td>
<td>0.225</td>
<td>0.379</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>0.256</td>
<td>0.384</td>
</tr>
<tr>
<td>$^{226}\text{Ra}$</td>
<td>0.0527</td>
<td>0.0779</td>
</tr>
<tr>
<td>$^{241}\text{Am}$</td>
<td>0.0529</td>
<td>0.0796</td>
</tr>
</tbody>
</table>

*In equilibrium with its decay products (till $^{210}\text{Pb}$ excluded)

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proportional to the number $N$ of contaminated items processed in a shift. In fact, only the first term of Equation 1 is proportional to the number $N$, the second term accounts for the contaminated items, waiting for the processing, stored close to the worker. In this connection, the reasonable hypothesis adopted is that only one contaminated item is waiting for the processing, because the others are either shielded by the first one or they are located at greater distance from the worker. Moreover, the effect of the waiting item is strongly dependent on the specific work task considered.

In Table 5, the effective dose received by the workers, divided by department and work task, owing to the manufacturing operations on one item (column $E$/item) is shown.

For a comparison, evaluated effective doses calculated using Equation 1, under the following assumptions, are also shown:

- processing of all items of a shift, with the hypothesis that all of them are contaminated (unreasonable hypothesis) (column $E$/shift);
- five contaminated items processed during a shift (column $E$/items, $N = 5$).

Table 6 can be useful for a comparison between effective doses received by workers when they manipulate one item contaminated with the four different radionuclides considered ($^{60}$Co, $^{137}$Cs, $^{226}$Ra and $^{241}$Am).

Taking into account that calculated doses (relative to 1.0 kBq g$^{-1}$) are proportional to the radionuclide concentration inside the coil, it is possible to scale

<table>
<thead>
<tr>
<th>Department</th>
<th>Work task</th>
<th>$N$</th>
<th>$T_{rem}$</th>
<th>$E$/item</th>
<th>$E$/shift</th>
<th>$E$/items</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>h per shift</td>
<td>mSv</td>
<td>mSv</td>
<td>mSv</td>
</tr>
<tr>
<td>Chemical pickling</td>
<td>Entering control point operator</td>
<td>25</td>
<td>6</td>
<td>0.10</td>
<td>0.76</td>
<td>0.21</td>
</tr>
<tr>
<td></td>
<td>Exiting control point operator</td>
<td>25</td>
<td>6</td>
<td>0.07</td>
<td>0.11</td>
<td>0.08</td>
</tr>
<tr>
<td></td>
<td>Generic operator</td>
<td>25</td>
<td>6</td>
<td>0.11</td>
<td>1.02</td>
<td>0.26</td>
</tr>
<tr>
<td>Entering coil storage area</td>
<td>Crane driver</td>
<td>50</td>
<td>6</td>
<td>&lt;0.003</td>
<td>0.15</td>
<td>0.02</td>
</tr>
<tr>
<td>Cutting line</td>
<td>Cutting line operator</td>
<td>10</td>
<td>5</td>
<td>0.13</td>
<td>0.85</td>
<td>0.45</td>
</tr>
<tr>
<td>Packaging line</td>
<td>Line operator</td>
<td>12</td>
<td>—</td>
<td>&lt;0.01</td>
<td>0.11</td>
<td>0.04</td>
</tr>
<tr>
<td></td>
<td>Packaging operator</td>
<td>12</td>
<td>4</td>
<td>0.03</td>
<td>0.33</td>
<td>0.14</td>
</tr>
<tr>
<td>External storage area</td>
<td>Lift truck operator (lateral loading)$^b$</td>
<td>80</td>
<td>5</td>
<td>0.06</td>
<td>0.13</td>
<td>0.06</td>
</tr>
<tr>
<td></td>
<td>Truck operator (front loading)$^b$</td>
<td>80</td>
<td>5</td>
<td>0.07</td>
<td>0.07</td>
<td>0.07</td>
</tr>
<tr>
<td></td>
<td>Store-man assistant</td>
<td>80</td>
<td>—</td>
<td>&lt;0.01</td>
<td>0.07</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>External position</td>
<td>Gantry crane driver</td>
<td>80</td>
<td>8</td>
<td>0.04</td>
<td>0.88</td>
<td>0.08</td>
</tr>
<tr>
<td>Unloading from transport</td>
<td>Store-man</td>
<td>80</td>
<td>8</td>
<td>0.04</td>
<td>0.88</td>
<td>0.08</td>
</tr>
</tbody>
</table>

$^a$Number of coils that a single worker can manipulate in a shift
$^b$On the lift truck, at an average distance of >10 m from the coil, without considering truck shielding

Table 6. Effective doses of external radiation, $E$/item (mSv), received by a worker during contaminated item manipulation (1.0 kBq g$^{-1}$) owing to the four radionuclides considered.

<table>
<thead>
<tr>
<th>Department</th>
<th>Work task</th>
<th>$^{60}$Co</th>
<th>$^{226}$Ra</th>
<th>$^{137}$Cs</th>
<th>$^{241}$Am</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>effective</td>
<td>dose</td>
<td>$E$ per item</td>
<td>(mSv)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chemical pickling</td>
<td>Entering control point operator</td>
<td>0.10</td>
<td>0.06</td>
<td>0.02</td>
<td>&lt;1E–04</td>
</tr>
<tr>
<td></td>
<td>Exiting control point operator</td>
<td>0.07</td>
<td>0.04</td>
<td>&lt;0.01</td>
<td>&lt;07E–05</td>
</tr>
<tr>
<td></td>
<td>Generic operator</td>
<td>0.11</td>
<td>0.06</td>
<td>0.02</td>
<td>&lt;1E–04</td>
</tr>
<tr>
<td>Entering coil storage area</td>
<td>Crane driver</td>
<td>&lt;0.003</td>
<td>&lt;0.002</td>
<td>&lt;0.001</td>
<td>&lt;3E–06</td>
</tr>
<tr>
<td>Cutting line</td>
<td>Cutting line operator</td>
<td>0.13</td>
<td>0.07</td>
<td>0.03</td>
<td>&lt;1E–04</td>
</tr>
<tr>
<td>Packaging line</td>
<td>Line operator</td>
<td>&lt;0.01</td>
<td>&lt;0.01</td>
<td>&lt;0.002</td>
<td>&lt;1E–05</td>
</tr>
<tr>
<td></td>
<td>Packaging operator</td>
<td>0.03</td>
<td>0.02</td>
<td>&lt;0.006</td>
<td>&lt;3E–05</td>
</tr>
<tr>
<td>External storage area</td>
<td>Lift truck operator (lateral loading)</td>
<td>0.06</td>
<td>0.03</td>
<td>&lt;0.01</td>
<td>&lt;6E–05</td>
</tr>
<tr>
<td></td>
<td>Truck operator (front loading)</td>
<td>0.07</td>
<td>0.04</td>
<td>&lt;0.01</td>
<td>&lt;7E–05</td>
</tr>
<tr>
<td></td>
<td>Store-man assistant</td>
<td>&lt;0.01</td>
<td>&lt;0.01</td>
<td>&lt;0.002</td>
<td>&lt;1E–05</td>
</tr>
<tr>
<td>External position</td>
<td>Gantry crane driver</td>
<td>0.03</td>
<td>0.02</td>
<td>&lt;0.006</td>
<td>&lt;3E–05</td>
</tr>
<tr>
<td>Unloading from transport</td>
<td>Store-man</td>
<td>0.04</td>
<td>0.02</td>
<td>&lt;0.008</td>
<td>&lt;4E–05</td>
</tr>
</tbody>
</table>
the ambient dose equivalent rate for a concentration $C$, different from $1.0 \text{ kBq g}^{-1}$, using the following scaling factor:

$$H(C) = H'(10) \frac{C}{10^3},$$

where:

- $H(C)$ = ambient dose equivalent rate, $H'(10)$ (mSv h$^{-1}$), at the new concentration $C$;
- $H'$ = ambient dose equivalent intensity, $H'(10)$ (mSv h$^{-1}$), calculated for a concentration of $1 \text{ kBq g}^{-1}$;
- $C$ = radionuclide concentration (Bq g$^{-1}$) in the contaminated coil (assuming the same size of the reference one).

Evaluation of the internal contamination

In order to evaluate the possible internal contamination of workers (committed dose), one has to consider that:

- the possibility of having radioactive substances released directly from contaminated steel is negligible, since the contamination is non-removable (this is true for scrap metal produced in coil cutting process too);
- radon inhalation does not provide a significant risk for workers because $^{222}\text{Rn}$ that could be released is diluted in the work environment air. The big volumes and the frequent air changes make the radon concentration be comparable with the natural background;
- during the chemical pickling process, the foil weight is reduced by $\sim 0.6\%$ (corresponding to 157 kg for the coils considered here); the total activity removed from the coil is $\sim 157 \text{ MBq}$, assuming a coil initial concentration of $1.0 \text{ kBq g}^{-1}$;
- the concentration of $1.0 \text{ kBq g}^{-1}$ is very high and superior to the mean values presented in Table 2;
- the inhalation dose coefficient of $^{241}\text{Am}$ is greater when compared with the coefficients for the other isotopes analysed, i.e. $2.7 \times 10^{-5} \text{ Sv Bq}^{-1}$ (absorption type M, 5 μm AMAD$^{(17)}$);
- the activity possibly released by the coil stays in the HCl dip and, to a large extent, it does not disperse in the working environment (chemical pickling cycle is closed, in order to preserve workers from HCl exposure);
- waste HCl re-treating and waste $\text{H}_2\text{SO}_4$ crystallisation processes take place in closed cycle;
- during cutting process, metal fragments are created, but they cannot be respirable, even when they have small dimensions;
- the contamination from macroscopic metal scrap is non-removable as for the coil itself;
- workers use gloves suitable for manipulation of metal objects.

From the previous considerations, it is possible to state that, in general, radioactive materials inside the coil do not provide a significant risk for internal contamination of workers, compared to external radiation exposure.

Nevertheless, in the case of a contamination by a source of $^{241}\text{Am}$, the committed dose could not be negligible or even bigger than the dose due to the external radiation. The committed dose strongly depends on the pickling plant design (containment systems) and a specific analysis has to be developed in each case.

Risks for workers

Let us stress that in the present evaluation the most conservative conditions are considered: a coil contaminated with $1.0 \text{ kBq g}^{-1} \text{ }^{60}\text{Co}$.

Under these assumptions, effective doses shown in Table 5 are lower than the European dose limits for the population (1 mSv y$^{-1}$), considering a maximum number of 10 contaminated coils handled per year. Dose approaches the limit when all the manipulated coils in a shift are assumed to be contaminated. In Table 6 the estimated doses for the other radionuclides are shown and are lower than the $^{60}\text{Co}$ one.

Except for the case of $^{241}\text{Am}$, the committed doses are mostly negligible compared to external radiation exposure.

In every case the radiation exposure risk for people staying at 50 m from the coil is negligible and widely $<1 \text{ mSv y}^{-1}$.

CONCLUSIONS

The fortuitous presence of radioactivity in scrap metals to be sent to the melting process is a reality and it is not possible to exclude it, even if it is quite unlikely.

Some conservative scenarios have been assumed considering possible radioactive source features (type of radionuclide and its concentration). It is important to underline that:

- the assumptions used in the simulation are sufficiently conservative to state that resulting evaluations are conservative for the considered radionuclides;
- doses can be scaled proportionally to the radioactivity level inside the contaminated coil or to the number of contaminated coils manipulated in a year.

The doses assessment for internal contamination and external radiation exposure shows that dose limits for the population are respected if the number
of contaminated coils is <10, considering 60Co and 226Ra as the radioactivity source. If the source is 137Cs the limit observance is assured for concentrations equal to or less than the European clearance levels, the dose limits observance for the internal contamination is not assured and have to be verified in the specific case.

These evaluations may be compared with the clearance levels proposed by the European Union(11–13) and in particular for clearance levels that appear in the first part of RP 122(4) (Table 7). Even more so, for coils contaminated with concentrations equal to or less than the European clearance levels, the dose limits observance is assured during chemical pickling process if radioactivity sources are 60Co, 137Cs, 226Ra and 241Am, even if the worker internal contamination due to 241Am would be better to be furthermore investigated.

REFERENCES


APPENDIX A

Calculations used in the simulations

\[ N_i \] being the total number of photons emitted, MCNP can calculate the differential fluence as the number of photons with energy between \( E \) and \( E + dE \) hitting an infinitesimal spherical surface, for emitted photon:

\[ d\Phi = \frac{1}{N_i} \Phi(E) dE \left[ \frac{\text{photons}}{\text{cm}^2} \right] \] (3)

The quantity \( d\Phi \cdot N_i \) is the differential fluence for a given number \( N_i \) of emitted photons.

To obtain \( d\Phi \) normalised for disintegration, \( d\Phi \cdot N_i \) has been divided by the number of occurred disintegrations \( (N_i/d) \). The quantity \( d \) is the average number of emitted photons during each disintegration and it is the sum of the relative emission probabilities of each photon with respect to father nucleus. Then the spectral fluence per unit of activity is found using the following equation:

\[ \frac{\text{photons}}{\text{disint.} \cdot \text{cm}^2} = \frac{d\Phi \cdot N_i}{(N_i/d)} = d\Phi \cdot \frac{d}{(N_i/d)} \] (4)

Integrating this quantity over the whole energy range, the fluence \( \Phi \) in that point has been calculated.
The derivative of the fluence over the time is obtained considering that

- the coil contamination \((C)\) is \(1.0 \text{ kBq g}^{-1}\);
- the coil mass \((M)\) is \(2.6 \times 10^7\text{g}\);
- the total coil activity \((A)\) is \(A = M \cdot C = 2.6 \times 10^{10} \text{ Bq}\).

Then

\[
\Phi = \frac{d\Phi}{dt}\left[\text{photons cm}^2 \cdot \text{s}^{-1}\right] = \Phi\left[\frac{\text{photons}}{\text{disint. cm}^2}\right] \cdot A\left[\frac{\text{disint.}}{\text{s}}\right]
\]

To estimate the ambient dose equivalent, \(H'\)(10), at a depth of 10 mm in the tissue, the ICRP\(^{(16)}\) conversion coefficients \(k(E)\) in pSv cm\(^2\) between fluence and the ambient dose equivalent have been used. MCNP interpolates the points with a double logarithmic function\(^{11}\). To obtain \(H'(10)(E)(dE)\) (the differential ambient dose equivalent per hitting photons having energy between \(E\) and \(E+dE\)) per unity of activity, the following formula has been used:

\[
H'(10)(E)dE\left[\frac{\text{mSv}}{\text{disint.}}\right] = 10^{-9}\Phi(E)dE\left[\frac{\text{photons}}{\text{disint. cm}^2}\right] \cdot k(E)\left[\text{pSv cm}^2\right]
\]

The ambient dose equivalent is obtained by an integration of the previous relation over the energy spectrum. The ambient dose equivalent rate \(H'(10)\) (mSv h\(^{-1}\)) results from the following equation:

\[
H'(10) = \frac{dH'(10)}{dt}\left[\frac{\text{mSv}}{\text{h}}\right] = 3.6 \times 10^3 \cdot H'(10)\left[\frac{\text{mSv}}{\text{disint. s}}\right] \cdot A\left[\frac{\text{disint.}}{\text{s}}\right]
\]