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Masterproef

Radiological characterization of hot cells and glove boxes as a preliminary research for the future dismantling

Promotor :
ing. Niels VANDEVENNE
dr. PHILIPPE DAMHAUT

Lowie Brabants
Scriptie ingediend tot het behalen van de graad van master in de industriële wetenschappen: nucleaire technologie

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Preface

In view of a possible career in the nuclear industry, I chose a master's thesis subject that was related to the dismantling of nuclear installations. I believe that dismantling projects will gain in importance in the nuclear landscape in the next years. As a result, a master's thesis topic within a dismantling project would make it possible for me to get acquainted with decontamination and decommissioning activities.

I would like to thank NIRAS for giving me the opportunity to work on the ONSF-site in Fleurus. Working on this nuclear site was a great opportunity for me to enhance my knowledge of the nuclear industry.

Before proceeding to the contents of this thesis, I would also like to thank a few people who supported me and made it possible to do my work on the ONSF-site in Fleurus.

First of all I would like to thank the ONSF-team for their assistance and time. In particularly, I am thankful to my promoters dr. Philippe Damhaut and drs. ing. Niels Vandevenne for their help and contributions to my master's thesis.

Furthermore, I would like to thank Reginald Coomans and Guillaume Vael for their help with the preliminary study of the decontamination.

Finally, a special word of thanks goes to my parents for giving me the opportunity to study and do my thesis in Fleurus. They also kept supporting and motivating me to accomplish my studies and master's thesis.

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List of abbreviations

ADC

Analogue to digital converter

BMB

Best Medical Belgium S.A.

FANC

Federal Agency of Nuclear Control

FWHM

Full width half maximum

HPGe

High purity germanium

IRE

Institute for Radioelements

MCA

Multi-channel analyser

ONDRAF/NIRAS

Belgian Agency for Radioactive Waste and Enriched Fissile Materials

RAW

Radioactive waste

ROI

Region of interest

Abstract

The installations of Best Medical Belgium S.A. were used for the production of radionuclides and are intended to be decommissioned. Because of the production of these radionuclides, radioactive contamination was spread over the entire production process. The main objective of this master's thesis will be to characterize the radioactive contamination and activated materials which are present in hot cells and glove boxes that were used for the production of the radionuclides. Next to the identification of the radionuclides present in the contamination, the disposal options of the activated materials will be discussed and a preliminary research is performed for the decontamination of the hot cells.

For the sampling of the contaminated parts, 59 swipe samples were taken on various walls of the hot cells and glove boxes. For the sampling of the activated materials, 16 samples were drilled. Although the sampling for both types of radioactive materials is different, the samples were all measured and characterized with a HPGe detector. The preliminary research for the decontamination was performed in consultation with experts present on the site.

The levels of contamination of the glove boxes and hot cells were the highest on the work sheet, and were lower on the side walls. The identified radionuclides in the contamination and activated materials were Na-22, Co-57, Co-60, Ag-108m, Ag-110m, Cd-109, Bi-207, Tl-204, Tl-202, Zn-65, Mn-54 and Ge-68/Ga-68. The origin of these radionuclides was also linked to the original production process. The final disposal of the activated materials and the glove boxes will be as category A waste. The preliminary study of the decontamination shows that the hot cells will first have to be decontaminated with a chemical procedure using 3 chemical agents, followed by sponge jet blasting.

Abstract – Dutch

De installaties van Best Medical Belgium S.A. werden gebruikt voor de productie van radionucliden en zijn bedoeld om te worden ontmanteld. Door de productie van deze radionucliden is er radioactieve contaminatie verspreid over het hele productie proces. Het hoofddoel van deze master thesis is om de radioactieve contaminatie en geactiveerde materialen te karakteriseren in hotcellen en handschoenkasten die gebruikt werden in het productieproces. Naast de identificatie van de radionucliden in de contaminatie zullen ook de eindopties van de geactiveerde materialen besproken worden en een voorstudie worden uitgevoerd over de toekomstige decontaminatie van de hotcellen.

Voor de staalnames van de contaminatie werden 59 wrijfstalen genomen op verschillende wanden van de hotcellen en handschoenkasten. Voor de staalnames van de geactiveerde materialen werden 16 stalen geboord. Hoewel de bemonstering van beide types van radioactieve materialen anders is, werden alle stalen gemeten en gekarakteriseerd met een HPGe-detector. De voorstudie van de decontaminatie werd uitgevoerd in samenspraak met experten die aanwezig zijn op de site.

De niveaus van de contaminatie zijn het hoogste op het werkblad en lager op de zijwanden. De geïdentificeerde radionuclides in de contaminatie en geactiveerde materialen zijn Na-22, Co-57, Co-60, Ag-108m, Ag-110m, Cd-109, Bi-207, Tl-204, Tl-202, Zn-65, Mn-54 en Ge-68/Ga-68. De oorsprong van deze radionucliden werd ook teruggekoppeld aan hun productieproces. De eindoptie voor de geactiveerde materialen is als categorie-A-radioactief afval. De voorstudie van de decontaminatie toont aan dat de hotcellen eerst gedecontamineerd zullen moeten worden met een chemische procedure bestaande uit 3 chemische producten, gevolgd door sponge-jet-stralen.

1 Introduction

1.1 The setting

As nuclear installations reach the end of their working term, the decommissioning of the facility is a required final step following the exploitation of the facilities. It is expected that decommissioning operations will gain importance in the Belgian landscape in the following decade due to the age of Belgian class I installations, such as the nuclear power plants. [1]

Due to this increasing importance of decommissioning, NIRAS, the Belgian agency for radioactive waste and enriched fissile materials, estimates that radioactive wastes (RAW) deriving from decommissioning operations will be the main source of RAW in the future. It is therefore crucial to perform decontamination and decommissioning with methods that minimize the amount of RAW and the associated economical costs. [2]

Next to class I installations such as nuclear power plants, there are also class II or III nuclear installations which cannot be neglected in terms of wastes. An example of such a class II installation that is currently being decommissioned are the infrastructures of a company named Best Medical Belgium S.A. (BMB). [3]

Best Medical Belgium was a nuclear company located at the nuclear site of Fleurus which produced radio nuclides mainly for applications in the medical industry. Normally the operators of nuclear installations are themselves responsible for the dismantling of their installations after the infrastructure has stopped exploitation. However, in certain specific situations such as the bankruptcy of BMB in 2012, NIRAS can be appointed for the execution of the preparatory work related to the dismantling and the dismantling itself. [4]

1.2 The problem statement

One of the core activities of BMB was the production of radionuclides for medical applications. The main medical nuclides which were produced at the site were I-123, Tl-201 and F-18. BMB also performed research and development on the production of Ge-68. Next to these medical radionuclides BMB also produced several other nuclides such as Co-57.

For the production of these radionuclides, BMB used two different cyclotrons: the first cyclotron was a CGR-cyclotron of 70 MeV, the second was an IBA cyclotron of 30 MeV. During the production process these cyclotrons were used to irradiate target materials. The irradiation of these targets induced nuclear reactions resulting in the formation of both the desired end product and several other side products. At the end of the irradiation, the targets would then be transported to the purification lab (chemical zone) where the produced nuclides were separated from the target materials and purified by a chemical process.

During the irradiations, the target materials as well as the cyclotron and the concrete walls of the vault were activated due to neutron and proton induced nuclear reactions. Because of the short range that protons can travel in materials, only components which were in direct contact with the proton beam, such as the target materials, underwent proton induced nuclear reactions.

Components which were not in direct contact with the proton beam were activated by neutrons which were released in the nuclear reactions. Next to the desired activation products, there were also other competing nuclear reactions which were responsible for the production of other undesired nuclides. Because of the transportation, chemical treatments and purification, these undesired activation products and contaminants were distributed and spread out over the entire production process.

In view of the future decommissioning it is essential to characterize the radioactive contamination of the installations of BMB. The characterization will provide information about the amount and type of RAW which will be produced as a result of the decommissioning. The characterization can also give information about the decontamination techniques that can be applied and the accompanying costs. Because of the extent of the contamination, this thesis will focus on the hot cells and glove boxes present in the chemical zone which played a vital role in the purification of the nuclides. The central theme of this thesis will be to characterize the radionuclides present in these hot cells as well as giving a preview of the possible decontamination techniques that can be used during the decommissioning of these hot cells and glove boxes. Figure 1 gives the line-up of the hot cells which are present in the chemical zone.

Figure 1: hot cells in the chemical zone

1.3 The objectives of the thesis

The most important objective of the thesis is to provide radiological information on the sort of radionuclides that are present in the hot cells and to provide a first indication of the decontamination techniques that can be used. Finding an answer to this problem involves the completion of an entire research process. In this process, several other objectives can be formulated.

The first objective is to perform a literature study in order to determine and select the possible materials and methods that can be used to characterize the radionuclides present in the hot cells. The literature study also involves gaining an understanding of the production process for the different nuclides. This understanding can give a preview of the different nuclides that can be found in the chemical zone as well as their spreading over the different hot cells. A different aspect of the literature study is obtaining information on decontamination techniques in order to provide a first indication of the decontamination techniques that can be used in the future decommissioning of the hot cells and glove boxes.

The second objective is to follow safety training on the site in order to be able to work in controlled areas and to work in a safe manner during the sampling process.

A third objective is the actual sampling process and the taking of representative samples. The locations of the samples will be carefully chosen in consultation with the staff of NIRAS. Prior to the sampling, an evaluation will be made in order to minimalize the doses received during the sampling process. After the sampling process, the samples will be labelled and measured.

The final objective is the correct interpretation of the data in order to provide an answer to the main objective which was mentioned earlier.

1.4 Materials and methods

For the literature research, different papers, books and libraries will be consulted. The libraries consist of both the physical library of the University of Hasselt (UHasselt), as well as the online data bases provided by the UHasselt and the Catholic University of Leuven (KU Leuven). For background information on the production process for the different nuclides, internal documents of BMB will be looked into. For additional information about the production process, the employees of NIRAS will be consulted.

During this thesis a distinction will be made between contaminated materials (such as the walls of the cells) and activated materials (such as the targets). This distinction is necessary because the samplings of activated and contaminated materials are different.

In order to measure the specific activity of the activated materials, drilling campaigns will be performed and the resulting drilling chips will then be measured. For the sampling of the contamination, swipe tests will be performed. The location of the samples will be chosen in cooperation with the employees of NIRAS and, as mentioned earlier, an evaluation will be made in order to minimalize the doses received during the sampling process.

Although the samplings of the activated and contaminated materials are different, the measurements for both samples are the same. The collected samples will all be measured with a high purity germanium detector (HPGe). After the measurement, the obtained spectra will then be analysed with Apex Gamma software.

The first chapter of this thesis will give an introduction about NIRAS and Belgoprocess and their function within the Belgian nuclear landscape. This chapter will be followed by a general chapter devoted to decommissioning and the project at Fleurus. Chapter four will focus on the chemical zone of building B14 and the hot cells and glove boxes that are present in this zone. This chapter will further elaborate on the functions of the hot cells and glove boxes. In order to understand the concepts of activation and contamination, an explanation of these concepts will be given in chapter five. The next chapter, chapter six, will be devoted to the production process of the radionuclides. The information of the historic production will be valuable later on in the thesis when the found radionuclides will be linked to the production process of their origin. The last theoretical chapter, chapter seven, will explain the future disposal scenarios of radioactive materials, which will be of importance when discussing the results and future destinations of the sampled materials. The methodology that was used for the sampling of the activated materials and the contamination will be discussed in chapter eight. Chapter nine will be devoted to the results of the measurements and the origin of the found radionuclides. The results of chapter nine will be compared with the disposal options of chapter seven in chapter ten. The preliminary study of the decontamination will also be a part of chapter 10. Finally, chapter 11 will give an overview of the conclusions of this master’s thesis.

2 ONDRAF/NIRAS and Belgoprocess

As long as the activity of radioactive waste (RAW) hasn’t decayed to acceptable and safe levels, all necessary safety precautions should be taken concerning the protection of people and the environment from the hazards of ionizing radiation. This commitment is at the core of the tasks of NIRAS, who’s responsible for the national management of radioactive waste in Belgium. The management of radioactive waste consists of tasks that are very diverse. [5]

The activities that the legislature has entrusted to this institution are the management of an inventory of all current and future radioactive waste, the search for new solutions concerning RAW and providing various services to companies such as collection, processing, conditioning and storage of their radioactive wastes. Furthermore, the institution has normative tasks such as recognitions of installations for processing and characterization of waste, the establishment of acceptance criteria and the approval of decommissioning plans for nuclear installations. [5]

NIRAS is the acronym for the national organization for radioactive waste and enriched fissile materials. The company was founded in 1980 and its duties were described in the Act of August 1980 and the Royal Decree of 1981. NIRAS is a public institution at the service of the community and is under the guardianship of the Minister of energy and economic affairs. [5]

The challenges which NIRAS faces with the management of RAW aren’t all of a technical nature but also include financial, ethical, ecological and social aspects. Therefore NIRAS always searches for sustainable solutions with respect to people and the environment now and in the future. [5]

NIRAS has developed a management system to ensure the protection of people and the environment against the dangers of radioactive waste. In order to guarantee the quality of the management system there are always two basic principles used, namely the containment of the radioactive material and the shielding of radioactive material (protection against ionizing radiations). The developed management system can be divided into the following five steps: [6]

- Step 1: limitation of waste, sorting and identification:
  The radioactive waste producers must limit the production of RAW as much as possible through recycling and recovery. The waste must also be sorted and then specified by content. [6]

- Steps 2-3: reduction of the volume, stabilizing and embedding:
  The objective of the conditioning is the reduction of the waste volume and to embed the radioactivity in barrels. The result is an end product that can be safely stored. [6]

- Step 4: storage of the waste:
  The barrels are then stored in buildings that are specially designed to protect against the harmful effects of ionizing radiation. Although this storage is safe for short and medium term, it is only a temporary solution. [6]

- Step 5: The long-term management:
  For the storage of radioactive waste in the long term NIRAS, is engaged in research into surface storage facilities and geological storage of RAW. [6]

The figure below illustrates the five step management system of NIRAS. In each of these steps the two basic principles are applied.

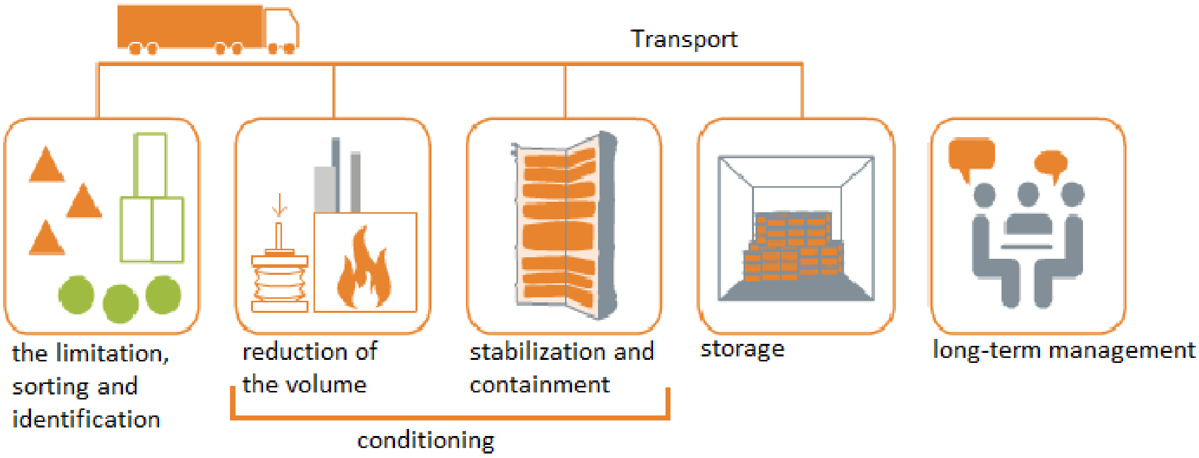

Figure 2: management system for radioactive waste [7]

For the conditioning process, NIRAS is assisted by its subsidiary Belgoprocess who has the necessary knowledge, experience and infrastructure for the safe processing, conditioning and storage of radioactive wastes and the decommissioning of nuclear installations. After the various radioactive waste materials are sorted, the volume is reduced in size as much as possible. Liquid radioactive waste is treated chemically or thermally and is reduced to a small volume of sludge. Solid combustible waste is incinerated whereas solid, non-flammable waste is super compacted into a disk of about 25 cm. Non-flammable and non-compressible waste is cut and collected in standard containers. [5]

The residue that is left over after the processing can then be encapsulated in glass, bitumen or cement so that the radionuclides are immobilized. Eventually everything is packed into drums and stored in the provided storage facility of Belgoprocess. [5]

Figure 3: NIRAS/ONDRAF [4]

3 Nuclear decommissioning

3.1 Financial aspect

One of the tasks which the legislator has entrusted NIRAS with is the responsibility of collecting and evaluating data on the decommissioning of Belgian nuclear installations, the approval of the decommissioning programs and the execution of these decommissioning programs in case of bankruptcy of the operators. [8]

Decommissioning costs belong to the owner of the nuclear facility. However, when the operators do not dispose of the planned financial resources needed for the decommissioning of the nuclear facility, NIRAS will be in charge of the decommissioning project and will look for potential financial responsibles. If no financial responsible is identified, the “insolvency fund” will be used to finance the project. Because the formation of a nuclear passive imposes the community with unreasonable financial burdens, NIRAS was appointed to make a five-year inventory consisting of the following aspects: [8]

- An inventory of all the nuclear installations in Belgium; [8]
- A cost estimate of the decommissioning of the nuclear installations;
- An evaluation of the existing and the adequacy of the financial resources needed for future decommissioning operations. [8]

This inventory is a means to prevent the creation of a nuclear passive because it enables the government to intervene in time to ensure that the operator has the necessary funds for the decommissioning operations. The figure below gives an overview of the nuclear facilities located in Belgium. [8]

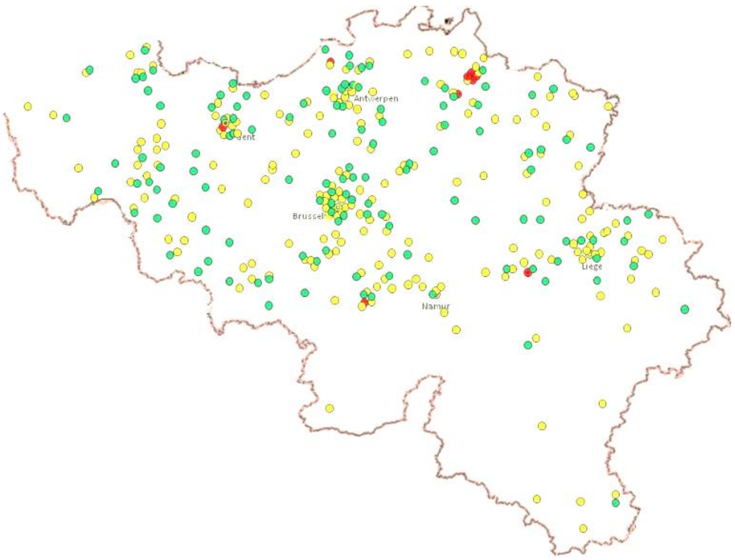

Figure 4: overview of Belgian nuclear installations; class I (red), class II (yellow), class III (green) [3]

The total number of nuclear installations in 2010 was 685 installations. 653 of these installations had a license and 32 sites had no licence. The 653 licenced installations contained 10 class I installations, 433 class II installations and 210 class III installations. The classification of the installations is a means to evaluate the required regulatory control and safety aspects of the installations as well as an indication of the required funds needed for future decommissioning of the installations. [3] [8]

Because of the bankruptcy of BMB, NIRAS was appointed for the decommissioning of the facilities. The installations in Fleurus are class II nuclear installations. [4]

3.2 Sanitation and declassification

When nuclear facilities reach the end of their operating time, they will inevitable need to be decommissioned. The sanitation and declassification are two key concepts in decommissioning. The main goal of the sanitation and declassification is to protect both people and the environment from harmful hindrance caused by the closed-down facility. [9]

- Sanitation consists of discarding radioactive components present in the nuclear installation prior to the decommissioning and declassification. [9]
- The goal of declassification is to exclude the installation of specific legislation concerning nuclear requirements. [9]

Declassification is the result of both administrative and technical procedures. Administrative obligations include the establishment and approval of decommissioning plans and the approval of release procedures. Technical procedures involve the practical sanitation, dismantling steps and the management of the wastes coming from the dismantling process. [9]

It is estimated that wastes deriving from the dismantling of nuclear facilities will be the biggest source of radioactive wastes (RAW) in the near future for Belgium. It is therefore crucial to use dismantling techniques which limit the amount of RAW. [9]

By using correct decontamination techniques, either chemical or mechanical treatments, it is possible to limit the amount of RAW because the techniques enable to remove the radionuclides from components and make it possible to reuse the components or to release the materials. [9]

Decontamination can limit the amount of RAW but there will always be a fraction that inevitably remains as RAW. This waste includes activated materials, not decontaminable components, residues coming from the decontamination process (secondary wastes), structural components and possible contaminated soil. [9]

3.3 Planning of the decommissioning

Since 1991, every Belgian nuclear installation is obliged to have a decommissioning plan. This plan is a conceptual study of the technical and economic aspects of the future decommissioning, including the finances of the decommissioning. The plan is drawn up by the operators of the facilities and is then approved by NIRAS. The main goal of this plan is to provide a guideline for the future decommissioning and provide a first estimate for the needed finances for the decommissioning. [10]

As mentioned earlier, the BMB installations were appointed in 2012 to NIRAS for the carrying out of the decommissioning. This required NIRAS to establish the new affiliate “ONDRAF – Fleurus Site” which is imposed with the decommissioning of the facilities of BMB. [4]

The decommissioning of the installations of BMB in Fleurus is a two-step process consisting of a sanitation step and the actual dismantling. It is estimated that the preparatory work concerning the sanitation will take place until 2016. The actual sanitation will follow the preparatory work and is estimated to be executed in 2016 and 2017. Similar to the sanitation, the dismantlement will also consist of a preparatory step (2015-2017) and the actual decommissioning (2017-2021). During the decommissioning process of the installations, NIRAS will be collaborating with AV-Controlatom and Tractebel Engineering. As already mentioned, the radiological characterization is an important step in the preparatory work concerning the decommissioning. [11]

4 Chemical zone of building B14

4.1 General information

Building B14 of the NIRAS site in Fleurus is a building used for the production of several radioactive nuclides used in the medical sector. The building was formerly operated by a medical company called Best Medical Belgium S.A. After their bankruptcy, NIRAS received authorization from the FANC for the cleaning and preparations prior to the dismantling process. [4]

In building B14 there are two cyclotrons which were used for the irradiation of target materials. The irradiated targets were then transported to the chemical zone of the building where the samples could be manipulated in hot cells, hoods or glove boxes. This thesis will focus on the hot cells and several glove boxes in the chemical zone. [12]

The figure below shows a drawing of the ground floor of building B14. The chemical zone and the cyclotrons are marked in black as well as the glove boxes and hot cells which are marked in red. [12]

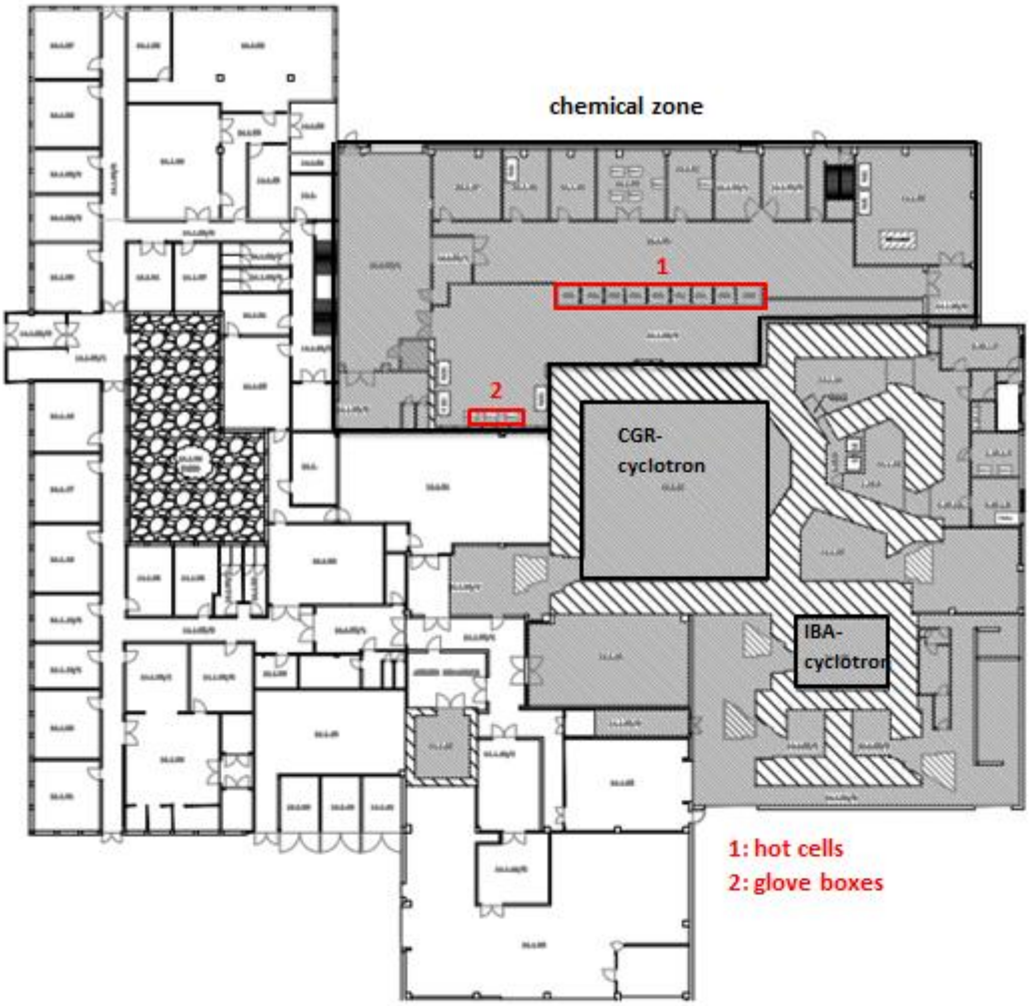

Figure 5: ground floor of building B14 [12]

The six hot cells present in the chemical zone of building B14 are called C30, C31, C32, C33, C34 and C35. Each of these cells had their own specific function in the production process of radionuclides. In addition to these cells there were also glove boxes and hoods. Three glove boxes will also be examined in this thesis. [12]

The general activities of the chemical zone were the purification of the irradiated targets coming from the cyclotrons present in the building, marking of molecules with the produced radionuclides, the conditioning of the products to obtain the desired end product and the initial phases of the packaging process. For operations with the most active materials the hot cells were used. For less active materials, the glove boxes and hoods were used. The following general rules were applied: [12]

- Manipulations of materials with activities in the order of one mCi ($37 * 10^6$ Bq) were performed under the hoods;
- The glove boxes with local shielding were used for manipulations with activities up to several hundred mCi ($37 * 10^8$ Bq);
- For more active materials, the hot cells were used.

4.2 Hot cells and their operations

The structure of each hot cell is approximately the same. They are all equipped with tele manipulators and lead glass. The outer layer of the cell walls and roof consist of lead. The thickness of the lead walls and roofing may vary from cell to cell as well as the number of tele manipulators. Production cells and packaging cells are equipped with 4 tele manipulators in contrast with sorting and connection cells which only have 2 tele manipulators each. Another difference is that each cell contains a stainless steel alfa box, except for the packaging cell. As mentioned before, the chemical zone contains six hot cells: C30, C31, C32, C33, C34 and C35. The row of hot cells and the content of cell C30 are shown in figure 6.

Figure 6: a) row of hot cells, b) content of cell C30

Sorting cell C35 is the first cell in the production process. The cell has two tele manipulators and its walls and roof consist respectively of 200 mm and 150 mm of lead. The conveyor belts connects the cell with the irradiation rooms of the cyclotrons. Cell C35 was used for the disassembly of the targets and their target mounts. The targets are then placed in plastic shuttles to transport them to the production cells C31 and C33.

Each production cell has 4 tele manipulators and the cell is divided into two areas which are separated with Lexan. The two areas of the production cells are a chemical zone and a distribution zone used to import or export materials to and from the chemical zone of the cell. The lead walls and roofs are slightly thinner and measure 150 mm for the walls and 100 mm for the roofs. These cells were used for the first chemical manipulations of the irradiated products.

In between each production cell there is a connection cell for the insertion and removal of materials. The two connection cells C34 and C32 have an airlock which has an opening to the front and rear of the hot cells making it possible to transport materials from the in and outside of the cell. Each connection cell has 2 tele manipulators and is built with the same lead shielding as the production cells.

4.3 Glove boxes and their operations

In the room at the back of the hot cells is the hot zone that contains three glove boxes. The three glove boxes played an important role in the preparation of the thallium targets. After each use, the irradiated targets were first left to decay until the activity fell below a certain level. The targets would then be transported to the first glove box where the surface of the target was cleaned with a sand blaster. The following step was performed in the second glove box. Here the targets were electroplated with a new layer of enriched ^{203}Tl target material. The final step of the production of the targets was the assembly of the target with the target holder.

After the treatment of the target materials in the glove boxes, the targets could be reused for another cycle of irradiation. Figure 7 shows the glove boxes.

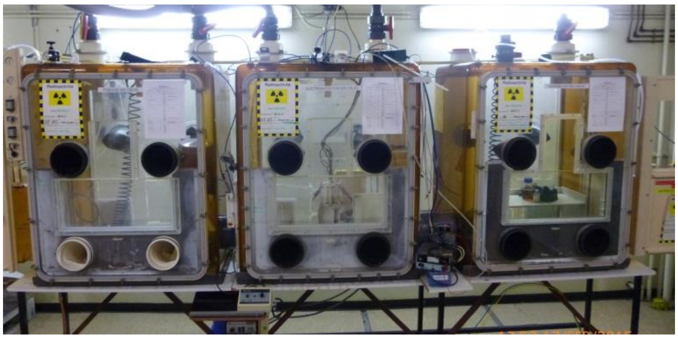

Figure 7: glove boxes

5 Activation and contamination

Understanding the concept of activation and contamination is an essential aspect to interpret the activation of the target materials and the origin of the contamination in the hot cells and glove boxes. Chapter 5 will therefore explain the principles of activation and contamination in more detail. The explanation of activation and nuclear reactions is needed to understand the nuclear reactions leading to the production of the desired radionuclides. The production of the radionuclides will be further explained in chapter 6.

5.1 Activation

A cyclotron is used to accelerate particles and bombard these particles onto targets. In these targets, the accelerated particles will induce nuclear reactions with the goal of producing radionuclides. A nuclear reaction can be described as a process where two or more nuclei interact with each other. The result of such a reaction can be a change in mass, energy or energy state of the resulting nuclides. A compact way of indicating a nuclear reaction can be the following: [13]

$$X\ (a, b)\ Y$$

X is the initial target nuclide at the start of the nuclear reaction with an accelerated projectile particle *a*. The interaction of *a* with nuclide X will form the reaction products Y and *b*. Typically, the nuclide Y is a heavier product that stops in the target, whereas *b* is a lighter particle. The resulting nuclide Y can be a stable nuclide or a radioactive nuclide. [13]

For the activation of the target materials and mounts, especially proton induced reactions will be of interest for the activation near the contact surface of the target with the proton beam. The proton type reactions are only relevant for the surface because the range of protons in the solid materials is in the order of a few micrometres. The neutron activation reactions will activate the targets further down the bulk of the material because of the longer range that neutrons can travel in materials. [14]

For proton induced nuclear reactions, a lot of different types of nuclear reactions can occur. The main reaction of interest were (p, xn) reactions resulting in the production of the desired nuclide on the target. Next to these (p, xn) reactions, other types of proton induced reactions are possible such as [15]:

- (p, γ)
- (p, pn)
- (p, d)
- (p, α)

P, γ, n, d and α stand for respectively protons, a gamma photon, a neutron, deuterium and an alfa-particle. All of these nuclear reactions can occur and depend on the irradiated target materials and the energy of the incident protons. The energy dependence of a (p, 3n) nuclear reaction is illustrated in Figure 8.

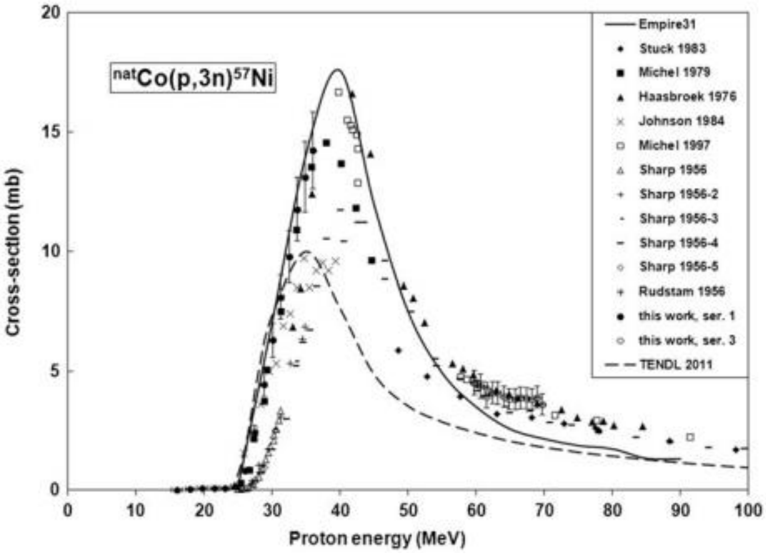

Figure 8: excitation function of the ^{nat}Co (p, 3n) ^{57}Ni reaction [16]

As can be seen in Figure 8, the cross section is strongly dependent on the energy of the protons. The minimum energy required for the (p, 3n) is about 15 MeV. At higher energies, the cross section increases rapidly to a maximum around 40 MeV. The energy dependence and the occurrence of all the competing nuclear reactions make it difficult to predict all the possible activation reactions. [16]

Next to the proton induced nuclear reactions, neutron induced reactions also have to be considered. The origin of the neutrons are the (p, xn) nuclear reactions. These neutrons can migrate further through materials without interacting because they are electrically uncharged. Just as in the case of protons induced reactions, various nuclear reactions are possible such as [15]:

- (n, p)
- (n, α)
- (n, γ)
- (n, xn)
- (n, f)
- (n, n’γ)

The cross section of neutrons for nuclear reactions is however different to the cross sections of protons with respect to the energy dependence. For neutrons, the probability of absorption usually increases with decreasing neutron energy of the incident neutrons. [13]

For an energy range from 0 to 500 keV, the most likely reactions are radiative neutron capture reactions (n, γ) resulting in an excited nucleus with an additional neutron. Neutrons with lower energies of few keV are more likely to undergo inelastic scatterings (n, n’γ). In this type of reaction, the same or a different neutron is scattered and a fraction of the energy is transferred to the nucleus resulting in an excited nucleus. For the emission of a different neutron, a minimum energy of about 0.1 to 0.5 keV is required. Neutron induced nuclear reactions which form charged particles, such as the (n, α) and (n, p) require higher energetic neutrons and gain importance from energies of 500 keV. The (n, xn) reactions can occur with even higher energies in the order of 10 MeV. [13]

Next to the variety of proton and neutron induced reactions, another important factor to consider is the variety of elements which are part of the target materials. In other words the chemical composition of the irradiated materials also has to be considered. The materials used to construct the target materials is not pure and consists of a number of other elements which can also be activated. Next to the chemical composition of the materials, the abundance of different isotopes of the chemical elements also has to be considered. It is also possible that the reaction products of previous nuclear reactions may again take place in further activation reactions.

In other words, making a prediction regarding all the possible nuclear reactions and thus making a prediction of all the possible activation products present in the activated targets is extremely complex and is not included in the scope of this study.

5.2 Activation sampling and free release levels

One of the goals during the dismantling process of nuclear installations is the minimization of RAW deriving from decontamination and dismantling procedures. In order to minimize the wastes, a possible strategy can be to free release as much material as possible which makes it possible to exclude these materials from legislation regarding radioactive materials. [17]

According to the Belgian law, materials that are present in controlled zones are considered as radioactive unless the materials are proven to be non-radioactive. The verification of the 'non-radioactivity' of the components includes the measurement of the radiological contamination/activation of the components. [1] [18]

An important factor for measurements that require sampling procedures is that the samples are representative with regard to the radiological aspects of the sampled material. For activated components this means that the sample must be taken in the bulk of the material at carefully chosen locations.

Next to the technical difficulties, legislative factors play an equally important role. This means that the samples have to be measured with an approved procedure in order to obtain valid results that are accepted by the legislature. This illustrates the importance of legislation with regard to the approval of measuring techniques. Next to the approval of measuring techniques, the legislator is also responsible for the release limits which are included in annex IB of the ARBIS. These limits determine the mass specific activity (Bq/g) limits below which a material can be exempted from regulatory control regarding their radioactive properties. The list of free release limits can be found in annex 4 of this thesis. It is also important to note that not all the radionuclides are included in this list and therefore it is possible that, when such radionuclides are found, agreements are made with the legislator. [18]

5.3 Radioactive contamination

Radioactive surface contamination is defined as unwanted radioactive material deposited in an uncontrolled manner on or in objects, in such concentrations that either operational inconvenience or radiological hazard is caused [19]. The contamination in the installations of BMB originates from the transportation of activated target materials, chemical treatments and purifications.

The total surface contamination is characterized as a combination of fixed and non-fixed or removable contamination. Fixed contamination is the fraction that is not transferred from contaminated surfaces to another uncontaminated surface when the two surfaces accidentally touch. The removable fraction can thus be seen as the fraction that can be easily transferred in normal working conditions. Unless information is available to indicate the contrary, it is assumed that the radiological properties are the same for both types of contamination and thus, the mix of radionuclides is the same for the fixed and non-fixed contamination. [20] [21]

The scope of this study will be to characterize all the radionuclides that are present in the hot cells and glove boxes and give an indication of the spreading of the contamination. The exact methodology that was used will be further elaborated in chapter 8. The sampling and measurements focus on the characterization of the non-fixed contamination in order to identify all the radionuclides that are present in the total contamination of the glove boxes and hot cells.

5.4 Contamination sampling and free release levels

As for activated materials, the sampling must also be performed to obtain representative samples. The contamination can be characterized with direct measurements, as well as indirect techniques.

Direct measurements of the contamination can only be carried out when the contamination is accessible with the used detector and when the measurement on the surface is not affected by the presence of other radionuclides. Another concern for direct measurements is that a possible contamination of the detector or operator can occur which has to be avoided. These two reasons are an important motivation for allowing the use of sampling procedures followed by indirect measurements. For the sampling of the non-fixed contamination, swipe samples will therefore be used. [20] [19]

Although the use of swipe samples for assessing removable contamination is considered to be a useful technique, several disadvantages have to be considered. One of the main concerns regarding the use of swipe samples for assessing removable contamination is the uncertainty on the collection efficiency. The fraction of the non-fixed contamination that is removed by a swipe sample depends strongly on the contamination mechanism, applied force, swipe material, wetting agent and the characteristics of the surface of the contaminated materials, such as the roughness or chemical composition. Figure 9 illustrates the collection efficiency for different swipe procedures. [21] [22]

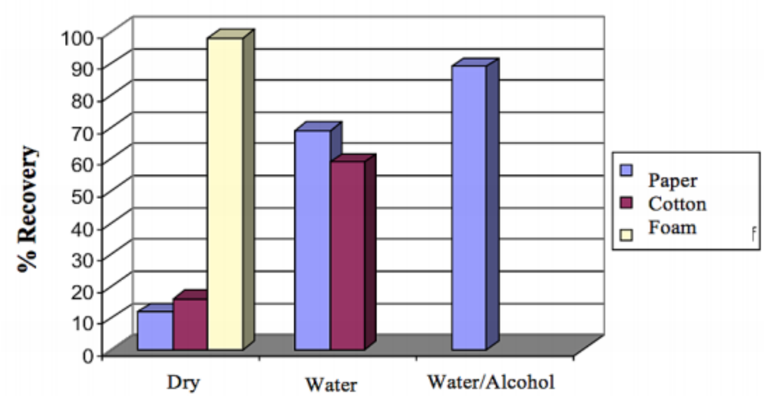

Figure 9: collection efficiency for different swipe procedures applied to a ^{32}P contamination [23]

As can be seen in Figure 9, the efficiency varies strongly with the used sampling procedure. For paper swipe samples the general efficiency is higher than the efficiency of cotton swipe samples. The efficiency also depends on whether dry or wet samples are used, and the used wetting agent. The used procedure for taking the swipe samples uses standardised paper swipe samples in combination with a soap solution. A general applied rule for estimating the swipe efficiency is a pickup factor or swipe efficiency of 20%. [23]

The aim of this study will be to identify all the radionuclides and give an estimation of the spreading of the contamination. In other words, the uncertainty on the pickup factor will not be of a great importance for the identification of the nuclides. A detailed explanation of the used procedure can be found further on in chapter 8.

Just as for the sampling of activated materials, the technical aspects of the measurements have to be recognised by the legislator. For surface contaminated materials, the release limits regarding the specific activity for exemption (annex IB of the ARBIS) have a lesser applicability to surface contaminated materials. For example, the majority of metal components that are being released are only surface contaminated. Limiting a material only by the mass specific activity can lead to surface contamination levels that are unacceptably high. Figure 10 shows this restriction by illustrating the relation between the maximum surface contamination and material thickness when only a mass specific clearance of 1 Bq/g is applied and thus no surface contamination limits are applied. [24]

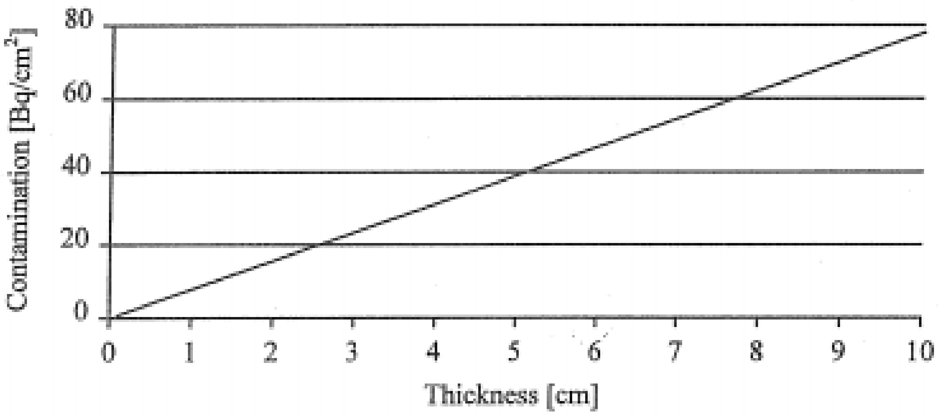

Figure 10: relation between maximum surface contamination and material thickness (1 Bq/g) [25]

As can be seen in the figure, the risk exists that thicker materials containing a high contamination level on the surface will still be considered as exempted from legislation if the mass-specific level is not exceeded. [25]

For materials which only have a contamination on the surface, clearance limits expressed in Bq/cm^2 could give a valuable additional clearance criterion next to the mass-specific limits. The IAEA basic safety standards define exemption levels for surface contamination at 0.4 Bq/cm^2 for beta and gamma emitters and 0.04 Bq/cm^2 for all alpha emitters. These values are however not included in the Belgian legislation as clearance limits and are only applied for transportation purposes. Unlike the Belgian legislation, the European Commission does describe radionuclide specific release levels for surface contamination. These limits can be found in RP114 concerning the definition of clearance levels for the release of radioactively contaminated buildings and building rubble. [25] [26]

6 Production process of nuclides

In order to make an estimation of the origin of the radionuclides present in the different hot cells and gloveboxes, it is important to give a general overview of the production process of the produced nuclides at BMB. The main nuclides that were produced by BMB in the hot cells are ^{123}I, ^{201}Tl, ^{57}Co and ^{68}Ge. In the next sections the production processes will be explained.

As mentioned in chapter 4, all the hot cells had their own function in the production process. Figure 11 illustrates all the nuclides which were handled in each hot cell. The figure only lists the desired nuclides which were handled in the hot cells. It is important to note that next to these nuclides, several other undesired nuclides (the side products) will inevitably be present in these hot cells.

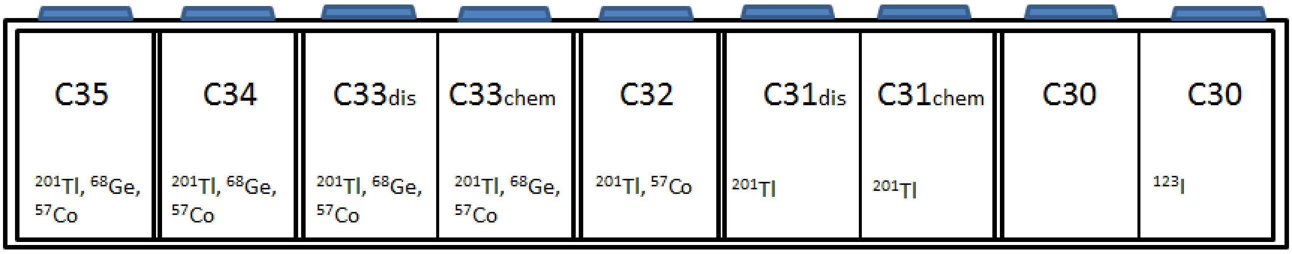

Figure 11: hot cells and the processed radionuclides

Hot cell C35 and C34 were used to handle the irradiated targets and therefore handled ^{201}Tl, ^{57}Co and ^{68}Ge. C33 was used for the distribution of the targets and the chemical purification of ^{201}Tl, ^{57}Co and ^{68}Ge. SAS cell number C32 only handled ^{57}Co and ^{201}Tl whereas C31 was used for the production of ^{201}Tl. The last cell, C30 was only used for the production of ^{123}I.

6.1 Thallium-201 production process

Thallium-201 is produced by the bombardment of an enriched thallium-203 target with protons coming from an IBA cyclotron present on the site. The ^{203}Tl is first electroplated onto a silver-plated solid target. After the target is prepared, it is ready to be sent to the irradiation room where the targets will be bombarded with 34 MeV protons for approximately 10 hours. The bombardment will result in the following nuclear reaction leading to the production of ^{201}Pb: [27]

$$^{203}\text{Tl (p,3n)}\ ^{201}\text{Pb} \text{ ---- (9.4 h) ---> } ^{201}\text{Tl (3.04 d)}$$

After the irradiation, the target is sent to the chemical zone of the building where the two-step purification process begins. The first stage involves the separation of the formed lead from the target material (containing ^{203}Tl) by washing the targets with a nitric acid solution. Fe^{3+} and ammonia is then added to the solution which forms a precipitate of iron and a co-precipitation of lead-201. The lead is then separated by filtration. The resulting precipitate is re-dissolved and the lead is extracted by using an anion exchange column. The resulting eluate is then evaporated and left to decay for approximately 30 hours. [27]

The second step extracts the thallium form the precipitate containing lead with an anion exchange column. After the second purification step, the Tl is now ready in the form of thallous chloride in a 0.9% NaCl solution. The operating conditions may vary depending on the specific activity requested by the customer. [27]

Next to the desired nuclear reaction there are also competing nuclear reactions which can give an explanation for nuclides present in the contamination of the hot cells. There are several possibilities such as nuclear reactions with other isotopes of Tl present in the target, nuclear reactions with nuclides present in the target other than Tl, or competing reactions with ^{203}Tl itself such as: [27]

^{203}Tl (p,4n) ^{200}Pb ---- (21.5 h) ---> ^{200}Tl (26.1 h)

^{203}Tl (p,2n) ^{202m}Pb ---- (3.62 h) ---> ^{202}Tl (12.23 d)

^{203}Tl (p, n) ^{203}Pb ---- (52.1 h) ---> ^{203}Tl (stable)

Because of the short half-life of ^{200}Tl, it is highly unlikely that this nuclide will be present in the contamination of the hot cells given that the production of ^{201}Tl was abandoned in 2009.

Next to the competing reactions, it is expected to find other activation products in the hot cells coming from the activation of other parts than the thallium coated part of the target. It is for example possible that the other materials in the target got activated. In order to make a prediction regarding the origin of the contamination, the activation and composition of the target is of importance. Figure 12 shows the front and rear view of the targets which were used for the production of ^{201}Tl. [27]

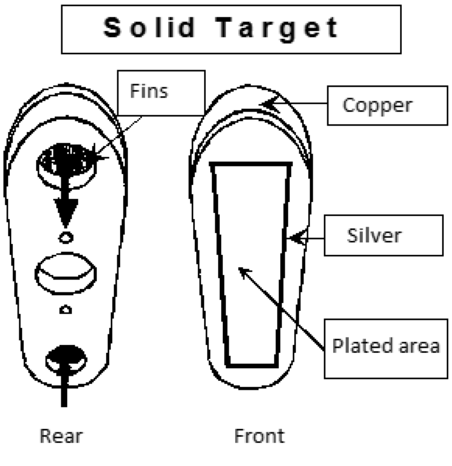

Figure 12: composition of the target used for ^{201}Tl production [27]

As shown in the figure, the target consists of copper and silver which is electroplated with ^{203}Tl. During the irradiation it is possible that not only the plated area is brought into contact with the proton beam coming from the cyclotron. The silver layer on the front of the target may as well be exposed to the beam resulting in possible activation caused by protons. Neutron activation will also occur, these neutrons are coming from the (p, xn) reactions in the ^{203}Tl plated area. The nuclides formed in this way could then be washed off the target and migrate throughout the production process. [27]

6.2 Iodine-123 production process

In contrast to the ^{201}Tl-production, the production of ^{123}I uses a gaseous target consisting of an aluminium alloy cradle filled with compressed ^{124}Xe gas. The target is irradiated for approximately 10 hours with a proton beam of 30 MeV. The irradiation will produce ^{123}Xe or ^{123}Cs which will both decay into the desired ^{123}I. The following reactions will occur: [28]

^{124}Xe (p,2p) ^{123}Cs ---- (5.8 m) ---> ^{123}Xe ---- (2.0 h) ---> ^{123}I

^{124}Xe (p,2p) ^{123}Xe ---- (2.0 h) ---> ^{123}I

At the end of the irradiation, the target is left to cool for at least 3 hours before the gas is pumped back into the storage vessel until the next irradiation. The target is washed and the washing material is pushed through a cation exchange column resulting in three separate fractions of eluate. The second fraction is the relevant fraction containing most of the activity. If the radioactive concentration of the fraction is too low, the solution can be evaporated until the desired concentration is reached. [28]

The end product is a sodium iodide aqueous solution in dilute sodium hydroxide. As with the production of ^{201}Tl there are also several competing nuclear reactions that may occur. These reactions are: [28]

^{124}Xe (p,3n) ^{122}Cs--- (4.2 m) ---> ^{122}Xe ---- (2.0 h) ---> I^{122}---- (3.6 m) ---> ^{122}Te (stable)

^{124}Xe (p, n) ^{124}Cs --- (31 s) ---> ^{124}Xe (stable)

^{124}Xe (p,4n) ^{121}Cs--- (126 s) ---> ^{121}Xe---- (39 min) -->^{121}I---- (2.12 h) ---> ^{121}Te--- (16.8 d) ---> ^{121}Sb (stable)

^{126}Xe (p, n) ^{126}Cs ---- (126 s) ---> ^{126}Xe (stable)

^{126}Xe (p,2n) ^{125}Cs---- (45 m) ---> ^{125}Xe---- (17 h) ---> ^{125}I (0.1 d)

^{126}Xe (p,3n) ^{124}Cs---- (31 s) ---> ^{124}Xe (stable)

^{128}Xe (p, n) ^{128}Cs ---- (3.62 m) ---> ^{128}Xe (stable)

^{128}Xe (p,2n) ^{127}Cs---- (6.2 h) ---> ^{127}Xe---- (36.3 d) ---> ^{127}I (stable)

^{128}Xe (p,3n) ^{126}Cs---- (1.64 m) ---> ^{126}Xe (stable)

^{129}Xe (p, n) ^{129}Cs ---- (32.2 h) ---> ^{129}Xe (stable)

^{129}Xe (p,2n) ^{128}Cs---- (3.64 m) ---> ^{128}Xe (stable)

^{129}Xe (p,3n) ^{127}Cs---- (6.2 h) ---> ^{127}Xe---- (36.3 d) ---> ^{127}I (stable)

Although there are many side reactions, it is not expected to find any of these products present in the hot cells or gloveboxes today due to their short decay times.

6.3 Cobalt-57 production process

The production of ^{57}Co used the same target material and setup as for the production of ^{201}Tl. The starting product for the production of ^{57}Co was an electroplated ^{58}Ni enriched target. The target was bombarded with a proton beam of 21 to 23 MeV for a period of approximately 3 to 4 hours. The following nuclear reactions produced the desired ^{57}Co: [29]

^{58}Ni (p,2n) ^{57}Cu--- (0.23 s) ---> ^{57}Ni ---- (1.5 d) ---> ^{57}Co

^{58}Ni (p, pn) ^{57}Ni--- (1.5 d) ---> ^{57}Co

After the irradiation, the targets were left to decay for 3 to 7 days before the ^{57}Co was further purified. The purification starts with the dissolution of the irradiation products by washing the targets with an HCl/HNO_3 solution. The following steps included elutions, precipitations and filtrations. [29]

The end product was cobalt chloride in a dilute HCl solution with a radio-purity of 99.6% ^{57}Co. Other isotopic side products in the production were ^{56}Co, ^{58}Co, ^{60}Co. Chemical impurities were Ni, Cu and

Fe. Co-56 can be produced by several reactions. The most relevant reactions are listed below: [30] [31] [32]

- ^{59}Co (n,4n) ^{56}Co;
- (n, t), (n, n+d), and (n,2n+p) reactions on Ni^{58};
- (p, p+d) and (p,n+2p)reactions on Co^{56}.

The nuclear reactions leading to the production of Co-60 will be discussed later in the thesis, in paragraph 9.6. The main nuclear reactions leading to the production of Co-58 are: [30] [31] [32]

- Ni^{58} (n, p) ^{58}Co ;
- (n,2n) and (p, n+p) reactions on Co^{59};
- Ni^{60} (p,n+2p) ^{58}Co;
- Ni^{61} (p,α) ^{58}Co;
- Ni^{62} (p,n+α) ^{58}Co.

6.4 Germanium-68 production process

Germanium was produced with a solution containing Ga-69 (p, 2n) and Ga-71 (p, 4n) mixed with nickel which was electrodeposited onto a copper target. The irradiations were conducted with a proton energy of 29 MeV. The two reactions responsible for the production of Ge-68 are: [33]

^{69}Ga (p, 2n)^{68}Ge

^{71}Ga (p, 4n)^{68}Ge

After chemical purification, the end product consisted of a Ge-68 and Ga-68 chloride solution. Important side products are Co-57, Co-56 and Ni-59. Ni-59 is produced by the following reactions; [33] [31] [32]

- ^{58}Ni (n, γ) ^{59}Ni;
- Co^{59} (p, n) ^{59}Ni ;
- (p,n+p) and (n,2n) reactions on Ni^{60}.

The nuclear reactions leading to the production of Co-57 will be discussed later in the thesis, in paragraph 9.6. The reactions leading to the formation of Co-56 were already discussed.

7 Disposal of radioactive wastes

As mentioned in chapter 5, the Belgian legislation contains release limits below which materials are considered to be non-radioactive. The disposal scenario of these materials is called an unconditional release.

When materials exceed these levels, they are considered to be radioactive leaving only two more disposal options possible. These two options are the disposal as radioactive waste or the melting of the metallic components at specialised melting facilities.

The most desired disposal option in terms of waste is the unconditional release because it produces no waste and enables the reuse or recycling of the materials. By contrast, the disposal as radioactive waste can be considered to be the most undesired option in terms of waste. Melting of the metallic parts can be seen as an intermediate option because during the melting, the radionuclides are distributed over the slags, bulk metal and the fly ashes. Because of this redistribution, it is possible that the activity of the resulting bulk is below the release levels. For this reason, the melting option can be situated in between the option of waste and release in terms of limiting radioactive wastes.

The limitation of the amount of radioactive wastes is however not the only decisive factor regarding the choice of the disposal scenarios. Other equally important factors are the associated costs and legislative restrictions.

7.1 Unconditional release

The criteria which a material must fulfil in order to be released are described in the ARBIS. Annex IB of this royal decree contains a list of radionuclides and their mass specific release levels. These release levels can also be found in annex 4 of this thesis. [18]

The release levels are based on a combination of 3 criteria which were used to compose the table of release limits (annex 4). These 3 criteria are: [18]

- the radiological risks to individuals are sufficiently low. More specifically the effective dose that may be sustained by any citizen per year, following the release of the materials is in the order 10 microsieverts per year or less;
- the collective radiological impact is sufficiently low. This means that the annual collective dose resulting from released materials has to be under a level of 1 man Sievert, and
- the risk of the occurrence of a situation that could lead to non-compliance with the criteria set out in the two previous points is negligible.

The release levels are only applicable when the materials were characterized with methods approved by the federal agency of nuclear control (FANC). For materials that contain multiple radionuclides, the following rule has to be applied: [18]

$$\sum_j (Cj \,/\, Cj,L) \leq 1 \qquad \text{with}$$

- *Cj* the specific activity of radionuclide j in the radioactive material
- *Cj,L* the release level for radionuclide j

In the case of a mixture of radionuclides and their decay products, decay products in this formula can be disregarded if they were taken into account when determining the clearance levels. Clearance levels which take into account the decay products are marked with a '+' in the table of release limits. [18]

7.2 Melting

For metallic components with activities that exceed the release levels, melting the metals could be an interesting technique to reduce the volume of radioactive wastes. Melting can be described as a volume reduction where the radioactive contamination is separated or partially separated from the bulk metal. Because of the melting the volume of the RAW is reduced and the radionuclides are redistributed over the slags, bulk metal and the fly ashes.

For the melting of radioactive materials, two main melting facilities exist. The first installation is located in Sweden and belongs to a company called Studsvik Nuclear AB. The second installation is owned by Energy*Solutions* in the United States (Utah). [34] [35]

7.2.1 Studsvik Nuclear AB

Studsvik Nuclear AB, located in Sweden, is a nuclear company that specializes is the treatment of radioactive wastes and consultancy services for the nuclear industry. For the treatment of low level contaminated and low activated metallic components, Studsvik disposes of melting facilities. In order to be able to meet quality and safety requirements, Studsvik uses several acceptance criteria for the metallic scrap metals. The acceptance criteria consist of both radiological aspects and non-radiological aspects. [35]

For the radiological aspects dose limits, surface contamination limits and specific activity restrictions are applied. The contact dose rate may not exceed 0.2 mSv/h, but smaller surfaces with a dose rates up to maximum 0.5 mSv/h can be allowed after acceptance from Studsvik. A second dose rate limit is also applied at a distance of 1 meter from the contaminated component. The dose rate at one meter may not exceed 0.1 mSv/h. Furthermore ^{60}Co is used as the dominating nuclide. For the contamination, a distinction is made between surfaces free of paint and painted surfaces (>50% of the surface is covered in paint). The following acceptance criteria applies for surface contamination: [35]

$$\frac{loose\ and\ fixt\ contamination\ (\frac{Bq}{cm^2})}{material\ thickness\ (mm)} < 10\ or\ 15$$

The limit of 10 is for contamination on unpainted surfaces, the limit of 15 is applied to painted contaminated materials. For alpha emitters the contamination may not exceed 10 Bq/cm^2 and for alpha emitting transuranium elements, the limit is more strict at 1 Bq/cm^2. Contaminated pipes are always to be discussed with Studsvik for a case-by-case approval. [35]

The specific activity restrictions apply the following acceptance criteria: [35]

$$\sum_{j} Cj < 50\ \frac{Bq}{g}\ or\ 100\ \frac{Bq}{g}$$

50 Bq/g is applied to beta and gamma emitting nuclides and 100 Bq/g is applied for alpha emitting nuclides. For slightly activated materials, the acceptance criteria of activated metallic scrap with a specific ^{60}Co activity exceeding 1.2 Bq/g have to be discussed with Studsvik for receiving a case-by-case approval. [35]

Next to the radiological acceptance criteria, Studsvik also applied non-radiological criteria to accept or decline waste. These criteria are: [35]

- Dimensions may not exceed 0.6 m in diameter/width and 1.2 m in length;
- No free liquids, grease, oil or spray cans;
- No toxic or hazardous components such as asbestos;
- Galvanised metallic scrap (zinc) is prohibited;
- Combustible materials that may self-ignite or cause explosion are not allowed;
- Lead composites (embedded in steel) are not accepted.

After the metallic materials are accepted, treated and melted the resulting ingots can be stored by Studsvik or can be sent back to the owner. Studsvik has the capacity to store annually around 20 tonnes of ingots. This storage is only used for ingots with activities that are expected to decay below the release limits within the maximum storage period of 10 years on the site of Studsvik. Other, more active ingots are sent back to the owner no later than 2 years after delivery to Studsvik. Other wastes deriving from the processing of the scrap metals (filters, slags…) are also sent back to the owner. [35]

7.2.2 Energy*Solutions*

The second facility that processes metallic scraps from the nuclear industry is Energy*Solutions.* As for the processing in Studsvik, Energy*Solutions* also uses acceptance criteria to evaluate if scraps can be accepted in their installations. These criteria are however slightly different from those used by Studsvik. [34]

For the dose rate, only metal scraps with a surface dose rate equal to or below 200 µSv/h will be accepted. The applied contamination limits are set at maximum 8 Bq/cm^2 for beta and gamma emitters and 1 Bq/cm^2 for alfa emitters. Unlike the specific activity limits of Studsvik, which are expressed in Bq/g, Energy*Solutions* uses a volumetric activity of 11 kBq/cm^3 that cannot be exceeded. This means that the accepted materials have a mass specific activity that depends on the density of the material. For stainless steel which has a density between 7480 and 8000 kg/m^3, dividing the volumetric activity level by the density yields a mass specific activity of 1471 to 1375 kBq/kg [36]. The volumetric activity of 11 kBq/cm^3 is applied to all radionuclides with half-lives longer than 5 years except for ^{3}H and ^{14}C. [34]

Unlike for the reprocessing at Studsvik, the bulk metals and the resulting waste and filters are not sent back to the owner. The resulting metal is reused in the nuclear industry. [34]

7.3 Radioactive waste

When waste materials exceed the activity limits for unconditional release or melting, the material can be classified as radioactive waste. In Belgium, conditioned radioactive wastes are classified in three groups: A, B and C. The distinction between the categories is based on the half-life of the waste and the contact dose rate. [37]

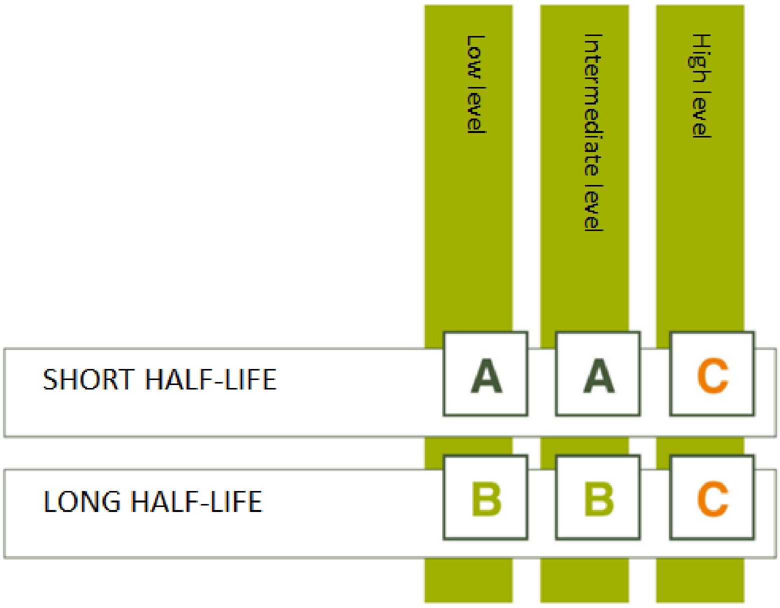

Figure 13: classification of radioactive wastes [37]

Radionuclides are classified as having a short half-life when the half-life is equal to, or lower, than 30 years. Radionuclides which have a half-life exceeding this period are considered to be long-lived radionuclides. For the classification on the base of the contact dose rate, the following classification is used: [37]

- Low level means that the contact dose rate does not exceed 5 mSv/h;
- Intermediate level wastes have a contact dose rate between 5 mSv/h and 2 Sv/h;
- The category of high level wastes applies to wastes with a contact dose rate higher than 2 Sv/h. An additional concern for this type of waste is the production of heat.

For the final disposal options of radioactive wastes of all three options, no solution is currently available. For the disposal of category A radioactive wastes, NIRAS has created the cAt project in Dessel. The cAt project aims to construct bunkers for the above ground storage of category-A radioactive wastes. [38] For the disposal of category B and C wastes, the option that is currently being considered is the long term geological storage in clay layers. [39] Figure 14 shows the plans for the future geological disposal and the cAt project. [38] [39]

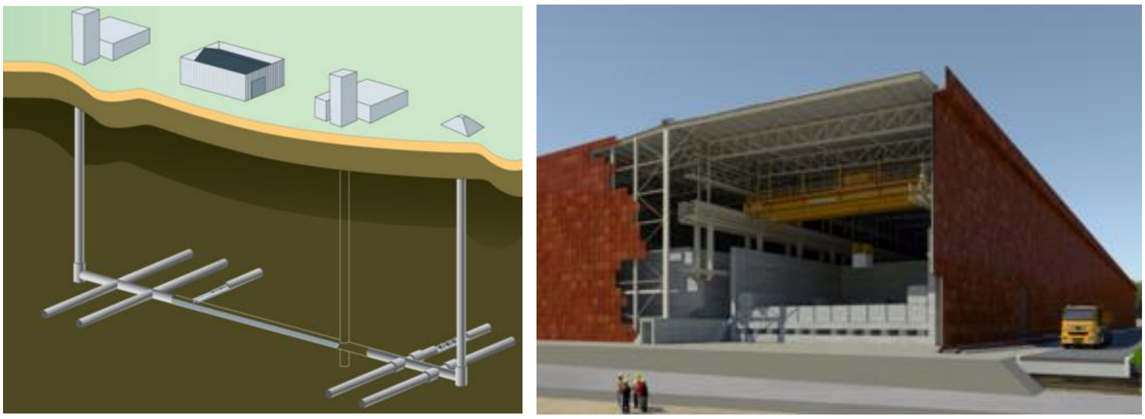

Figure 14: geological disposal (left), cAt project (right) [38] [39]

8 Methodology

Chapter 8 focusses on the methodology, techniques and the sampling procedures which were used in order to characterize the hot cells and glove boxes. As was mentioned earlier, the hot cells and glove boxes still contain a collection of materials. These materials consist of activated components such as the targets and the target mounts, and contaminated materials. The contaminated materials will be considered as waste because it is not worth the effort to decontaminate each piece one at a time. It is therefore not necessary to fully characterise these materials for the prospect of possible release. In contrast to the smaller contaminated materials, the contamination of the larger components will be considered important to characterise because these larger components are worthwhile to decontaminate in order to reduce the volume of produced RAW in the decommissioning process. The sampling of the contaminated parts will be discussed in the first paragraph of chapter 8 (8.1).

The second paragraph, chapter 8.2, will elaborate on the sampling of the activated materials. It will discuss the locations of the samples and the used techniques in order to collect representative samples.

Paragraph 8.3 will discuss the preparatory steps for the measurement of the samples and the geometry.

Finally, paragraph 8.4 will further focus on the principles of gamma spectroscopy, the specifications of the high purity germanium (HPGe) detector and the calibration of the HPGe detector.

8.1 Sampling of contaminated parts

All the materials and side panels of the glove boxes and hot cells can be considered to be contaminated. For the measurements of contaminated surfaces swipe samples were taken on all the side panels of the hot cells and glove boxes and their worksheets.

Because of the lack of standardisation of the technique on taking swipe samples an approved procedure for the collection of swipe samples was used. This procedure was established by AV Controlatom and approved by ONDRAF for the use at the Fleurus site. [40]

The procedure for taking swipe samples has the following steps: [40]

1. A standard type filter with a diameter of 60 mm is used for the sampling. The filter is humidified with an aqueous solution containing a regular detergent;
2. Swipe the region of interest on a surface of 300 cm^2. If swiping on an area of 300 cm^2 is not possible, a minimum surface of 100 cm^2 can be used as an alternative;
3. Place the swipe sample in a plastic bag and seal the bag;
4. Give the sample the correct name tag. The sample is then ready to be measured.

The swipe setup and the standard filter are shown in Figure 15. The use of the cylinder will be discussed further on in the text.

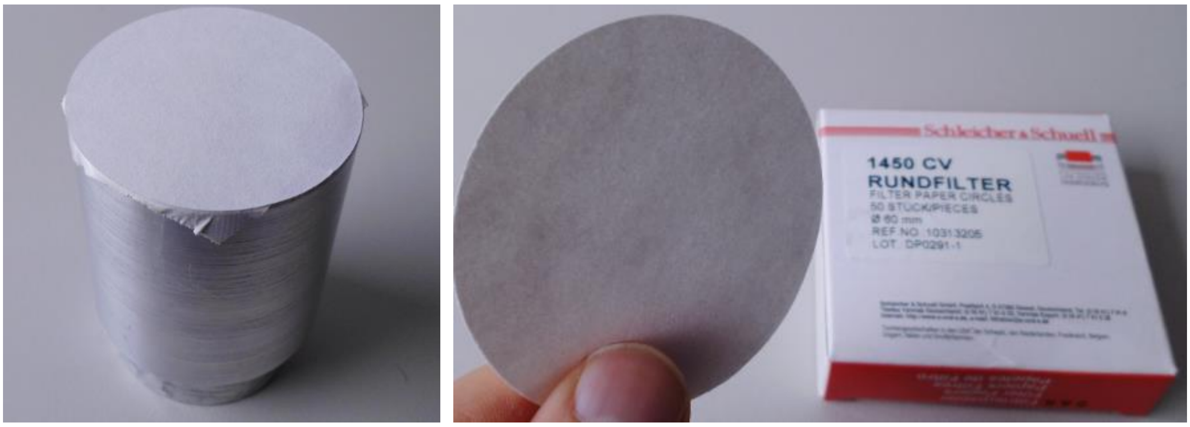

Figure 15: swipe setup and filter

Next to the use of an approved procedure for the collection of the samples additional measures were taken in order to enhance the accuracy and the reproducibility of the sampling. An example of such a measure is the use of a frame which defines an area of exactly 300 cm^2. Furthermore, the swipe filters were also mounted on a plastic cylinder. The weight of the cylinder was then used for applying always the same pressure to the swipe sample. The same cylinder was always reused in order to improve the reproducibility of the applied swiping pressure in comparison with manually taken swipe samples.

Swipe samples in the hot cells were taken using the tele manipulator arms of the hot cells. The other swipe samples were taken manually. Figure 16 shows the procedure for the sampling in the hot cells.

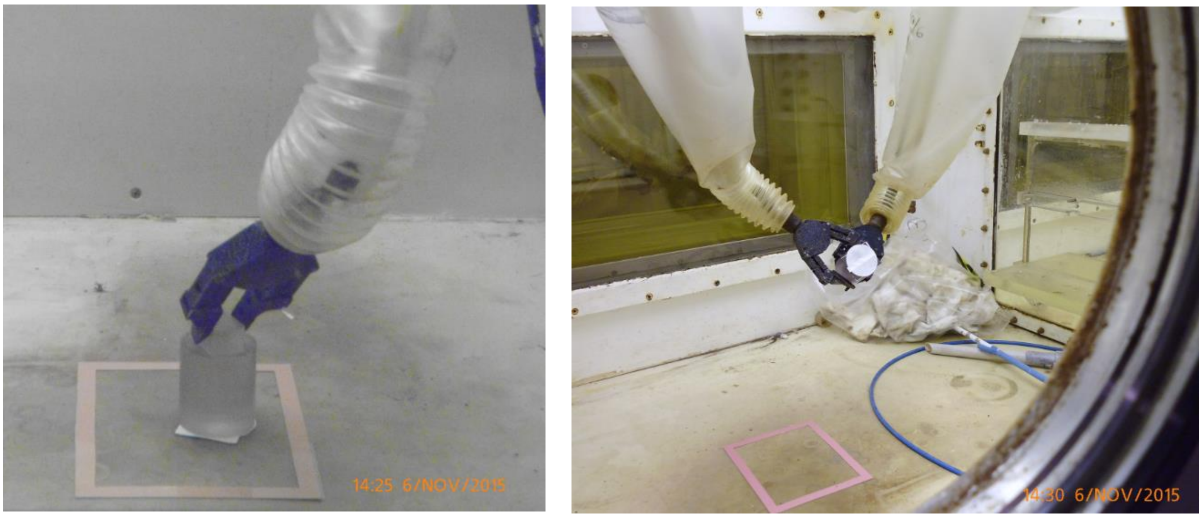

Figure 16: sampling procedure of the swipe samples

After the swiping, the samples were taken out of the hot cells through holes in the backside of the alfa box and placed in plastic bags. The samples are then ready to be measured with the high purity germanium detector.

8.2 Sampling of activated components

The activated materials were stored at sorting cell C35. The first step, preliminary to the actual sampling and drilling, was the removal of several targets and target supports from the hot cell. Because of the radioactive contamination in this cell, special care was taken in order to prevent the spreading of the contamination when the cell was opened.

The targets were picked up with the tele manipulator arms and were passed through a hole at the back side of the cell. The targets were then placed in a plastic bag and were sealed off with tape. As can be seen in Figure 17(left), hot cell C35 contains a lot of targets and target materials. Figure 17(right) shows a target mounted on a target mount.

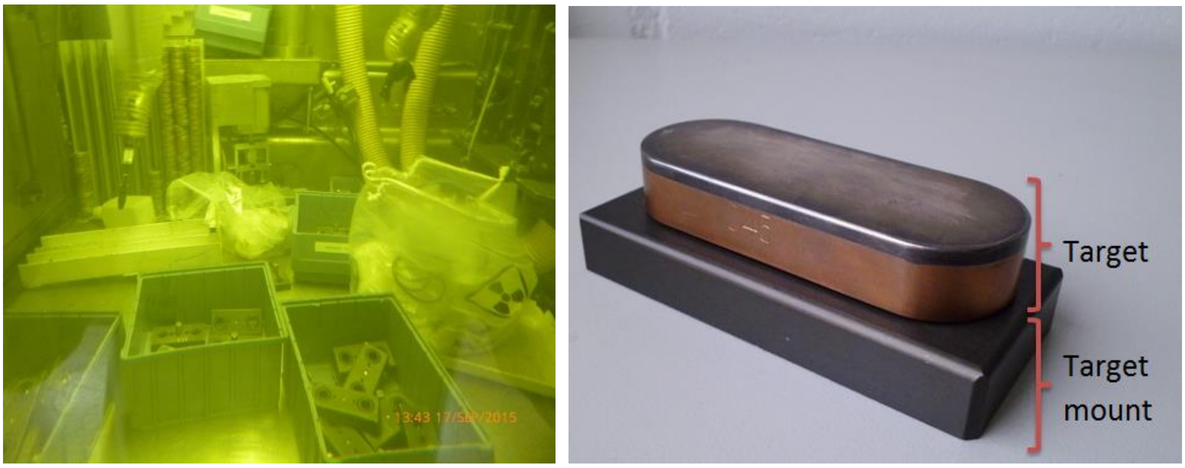

Figure 17: interior of C35 (left), target with target mount (right)

Due to the large number of targets and mounts in the hot cell, only a few were removed from the hot cell. The targets located in C35 are a combination of targets used for the production of Co-57 and targets for the production of Tl-201. The targets for both production processes are identical except for the engravings on the sides of the targets. Tl-201 targets were engraved with a number, Co-57 were engraved with “CO” followed by a number.

Because the irradiations for the production of Co-57 and Tl-201 were different in both energy and irradiation time, it is possible that there is a difference in the activation of the targets. In contrast to the targets, the mounts were identical for both productions and are indistinguishable because the mounts do not have different engravings. This means that a mount could be used for both productions.

Because of the difference in the Tl-201 and Co-57 targets and the similarity of the target mounts, the following series of samples were selected and removed from the hot cell:

- 4 Co-57 targets
- 6 Tl-201 targets
- 7 universal target mounts

The contact dose rate of these samples was then measured with a radiagem 2000 dose rate and survey meter in order to draw up an interval ranging from the least activated target to the most activated target. The list of all the measured targets and target mounts can be found in appendix 1.

Of all the activated materials, the most and least active of each interval were chosen for further sampling.

After the targets and mounts were selected, swipe samples were taken of the surface of the activated materials in order to characterize the removable contamination on the surface of the targets. The targets and mounts were then drilled with a vertical drill. The locations of the sampling were carefully chosen in order to be able to measure all of the activation products present in the targets and mounts.

The targets for the production of Co-57 and Tl-201 consist of a copper tablet with a surface of silver. The silver and copper parts of the target are welded together with a tin weld. Because different metals have different activation products, drilling samples were taken of each metal present in the target. The target mounts consist of an aluminium tablet with an inox support. Figure 18 shows the sampling points on the target and the target mount.

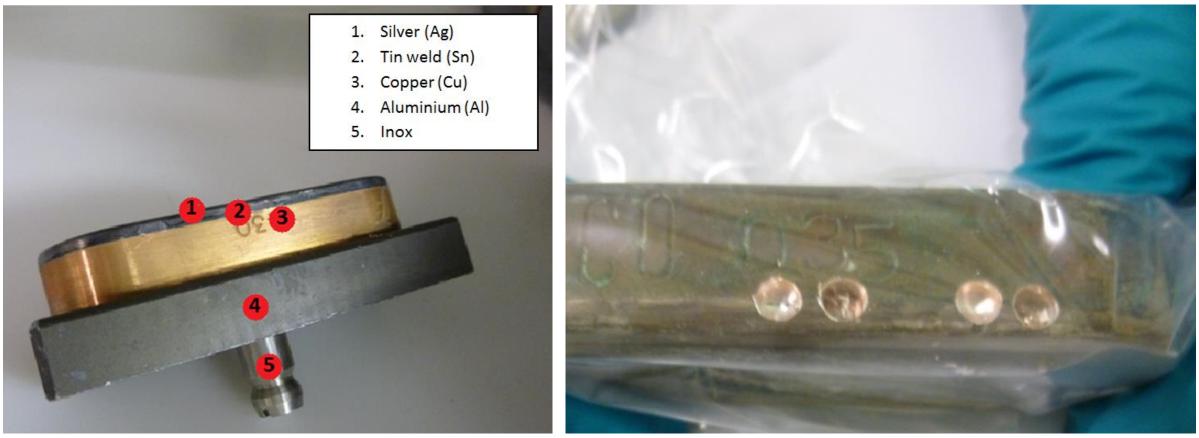

Figure 18 : sampling points (left), drill holes in target 'CO 035' (right)

For the targets (both Co and Tl), the least and most activated targets were chosen for further sampling. Each target was drilled in the silver, copper and tin part resulting in 3 samples for each target. As for the targets, the most and least activated mounts were chosen for further sampling. Because the mounts only consist of aluminium and inox only two samples were taken for each mount. Although the activation is dependent on the depth in the material, the mounts and targets were not drilled at various depts. Table 1 gives the production process, the type of activated material, the number and the amount of samples which were taken from the selected target or target mount.

Table 1: selected targets and target mounts with the production process

production process	type of activated material	number	amount of samples
Tl-201	target	905	3
		1034	3
Co-57	target	35	3
		22	3
Tl-201, Co-57	mount	A11	2
		B14	2

As mentioned earlier, the samples were drilled with a vertical drill. The activated target or target mount were placed in a plastic bag in order to capture all the drilling chips. The drilling setup is shown in Figure 19.

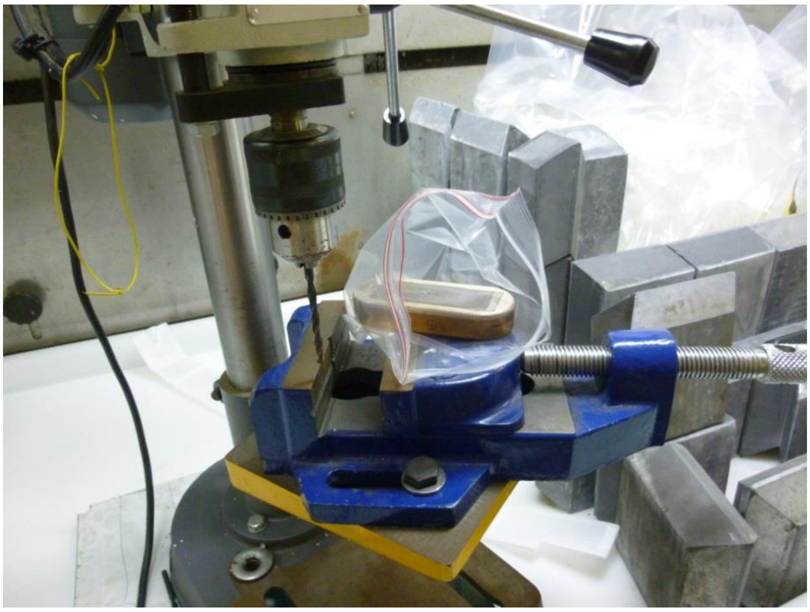

Figure 19: vertical drill setup

In order to avoid cross contamination of the samples, the targets and mounts were drilled with a series of different drill bits. The drilling chips were also kept in separate plastic bags to avoid cross contamination. Figure 20 shows the used drill bits and the drill chips.

Figure 20: drilling chips (left) and drill bits (right)

8.3 Measuring geometry

After the targets and mounts were drilled, the drill chips were put into a fixed geometry before they could be measured in the HPGe detector. The fixed geometry made it possible to measure all the samples the same way and thus making it possible to compare the results of the obtained drill samples. The geometry consists of a petri dish, a piece of PVC tube and a piece of foam. Figure 21 shows the used geometry for the measurements with the HPGe detector.

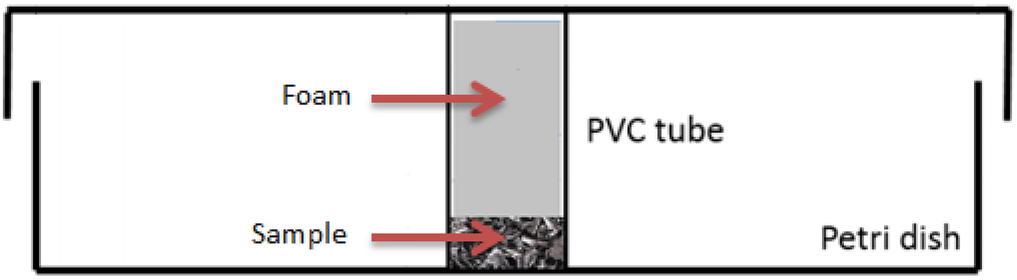

Figure 21: measuring geometry

The piece of PVC tube was glued to the base of the petri dish and filled with approximately 1 g of drilling chips. The drilling chips were then covered with a piece of foam to keep them in place. The lid of the petri dish was then glued to the PVC tube and the base of the petri dish. Figure 22 shows a photo of the used petri dish geometry.

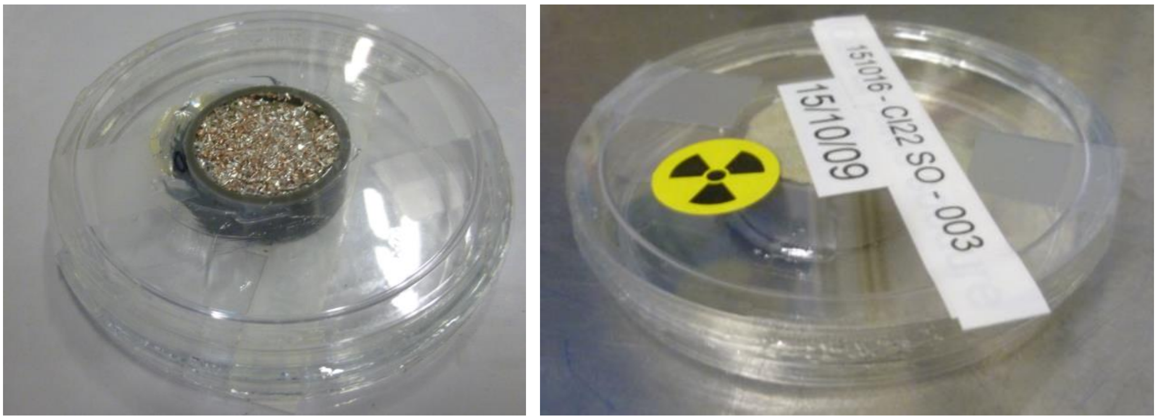

Figure 22: photo of the top and bottom side of the petri dish geometry

For the measurement of the contaminated surfaces, the swipe samples were placed in a plastic bag and were measured in direct contact with the detector. Figure 23 shows a swipe sample ready for the measurement with the HPGe detector.

Figure 23: example of a swipe sample

8.4 Gamma spectroscopy

8.4.1 General principle of gamma spectroscopy

Gamma ray spectroscopy is an analytical technique used for the identification and quantification of gamma emitting nuclides. The measurement gives a spectrum, the pulse height of which is proportional to the activity of the nuclide and the position on the horizontal axis gives the energy of the incident photons. [14]

The principle of the detection is based on the interaction of photons with materials. There are three ways in which a photon can interact with surrounding materials: [14]

- Compton scattering;
- Photo-electric absorption;
- Pair production.

The identification of nuclides is based on the measurement of peaks with energies equal to the total energy of the incident photons. In order to measure such a full energy peak, the total energy of the photon has to be deposited within the detector area. The following events give rise to full energy peaks: [41]

- Photo-electric absorption of a photon in the detector area;
- One or more Compton scatterings where the photons stay within the detector and finally undergo photo-electric absorption;
- Pair production followed by the photo-electric absorption of both annihilation photons within the detector;

For a theoretical detector with an infinite volume, all the energy of the incident photons would be deposited within the detector area because no energy could escape form the detector. The resulting spectrum would then only show full energy peaks in the spectrum. [14]

A more realistic detector also shows other events than full energy peaks such as a Compton continuum, escape peaks, sum peaks and influences from the detector environment. The Compton continuum is formed when scattered photons, coming from Compton scattering, escape form the detector taking with them a fraction of the initial photon energy. Pair production can occur when the energy of the photons is larger than two times the resting energy of an electron. Pair production results in the formation of an electron and a positron. Formed positrons due to pair production will then annihilate and form two annihilation photons. Escape peaks can then be observed when one or both of the photons escape the detector. These events can be seen in the spectrum as a single or double escape peak. The interactions of photons in the detector as well as a typical gamma spectrum are shown in Figure 24. [14]

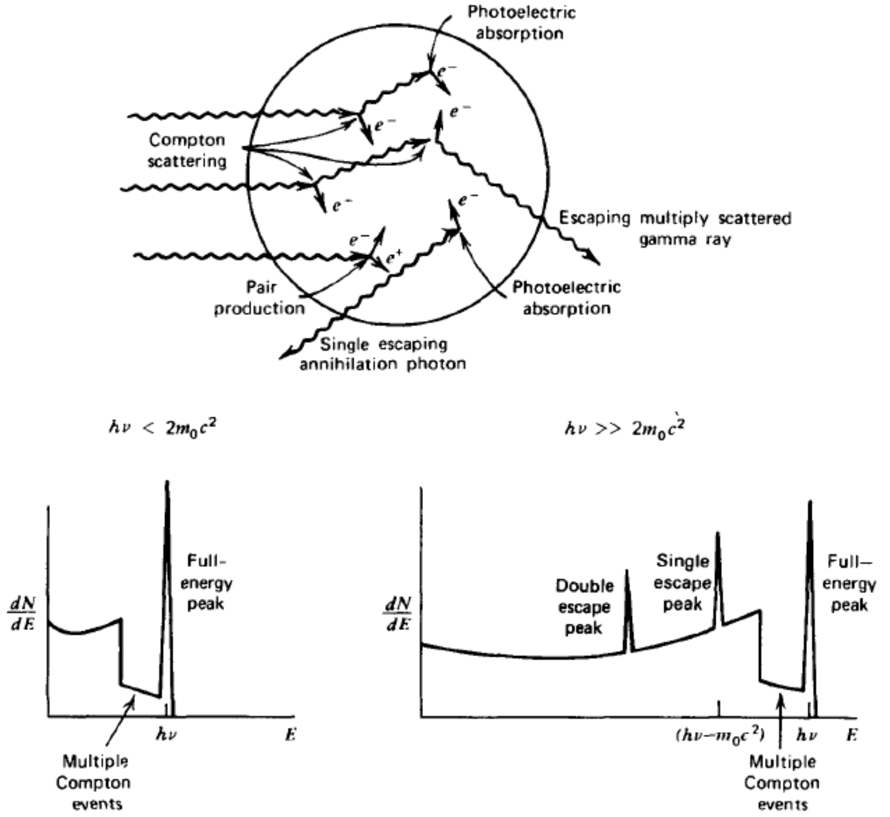

Figure 24: interactions of photons in the detector and the resulting spectrum [14]

When two gamma rays simultaneously deposit their energy in the detector, a sum peak can be observed at a channel with the combined energy of the two photons. The resulting peak may not be considered as a characteristic gamma ray of the radionuclide. The sum peaks have to be taken into account during gamma spectrometry. [14]

Photons can also interact with surrounding materials of the detector. These interactions result in secondary particles entering the detector and can thus be measured in the gamma spectrum. The interactions with the surrounding materials are characteristic X-rays from the detector shielding, annihilation photons coming from annihilations outside of the detector crystal and back scattering of photons. Figure 25 shows the interactions of photons with surrounding materials and the effects on the gamma spectrum. [14]

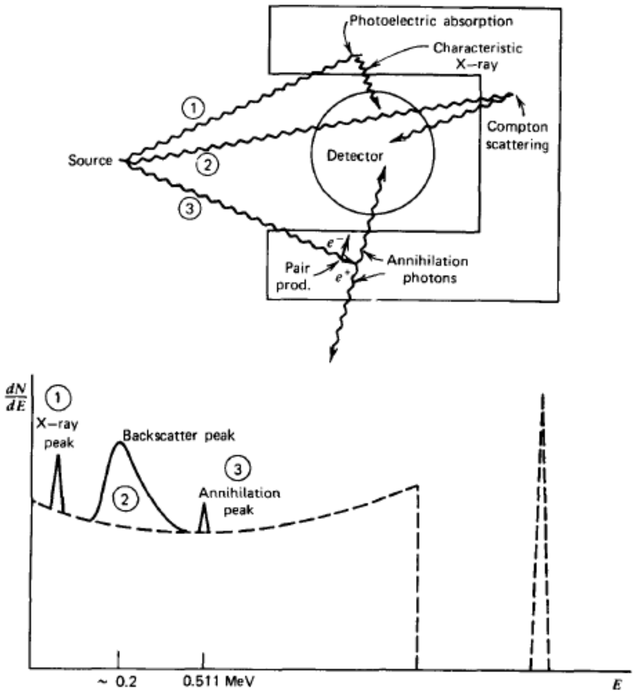

Figure 25: interactions of photons with surrounding materials and the results on the spectrum [14]

8.4.2 Detector specifications

For the measurement of the samples with gamma spectroscopy, a high purity germanium semiconductor detector was used. More specific, a coaxial germanium detector (type GX 2518) was used with a thin-window contact which extends the sensitive energy range down to 3 keV. Table 2 shows the detector specifications of the used GX 2518. [42]

Table 2: detector specifications [42]

Model number	Relative efficiency (%)	Full with half maximum (FWHM)			Peak to Compton ratio (P/C)	peak shape (FWTM/FWHM)	endcap diameter (mm)
		At 22 keV	At 88 keV	At 1,3 MeV			
GX 2518	25	0,7	0,8	1,8	54:1	1,9	76

The detector setup consists of the detector itself as well as a high power voltage supply of 3000 V, a preamplifier, an amplifier, an analogue to digital converter (ADC) and a multi-channel (MCA) analyser. [43]

The software used to measure the spectrums and analyse the results was Apex Gamma software 2000 from Canberra Industries. Figure 26 show the detector setup of the series GX 2518 HPGe-detector. [43]

Figure 26: detector setup

8.4.3 Calibration

Before starting measurements, an energy-calibration has to be executed in order to assign the appropriate energy to each channel. This energy-calibration has to be frequently performed because detector response is not absolutely stable under all conditions. In order to prevent an energy drift, the detector was calibrated each day of measurements. [41]

The goal of the energy calibration is to assign a correct energy to each channel. This is done by measuring the spectrum of a source with known energy peaks. A least square fit is then made which gives the equation linking the energy with the correct channel number. The source for the energy calibration was a Eu-152 source. This source has numerous peaks in a wide energy range, and is therefore an excellent source for energy calibrations. Table 3 shows an example of an energy calibration with the Eu-152 source. [41] [43]

Table 3: Eu-152 energy calibration [43]

Peak number	Energy (keV)	Channel	FWHM (Ch)
1	121,8	226,1	1,69
2	244,7	528,6	2,07
3	344,3	741,2	2,21
4	411,1	883,9	2,99
5	444,0	954,1	2,78
6	778,9	1669,2	3,07
7	867,3	1858,1	3,65
8	964,0	2064,6	3,42
9	1085,8	2324,8	3,55
10	1112,0	2380,7	3,56
11	1408,0	3013,0	3,98

Figure 27 shows the spectrum of the Eu-152 source and the corresponding calibration curve. An example of a calibration that was performed had the following equation for the least square fit:

$$\text{Energy} = -2.893 + 0.468 * \text{Ch} - 4.83\text{E} - 08 * \text{Ch}^2$$

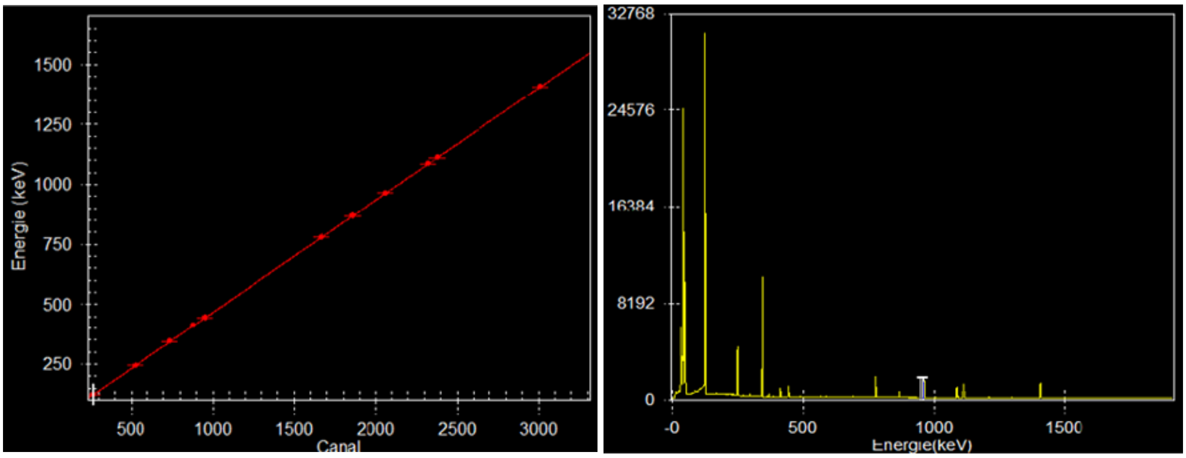

Figure 27: spectrum of the Eu-152 source and calibration curve

For the measurement of the specific activity, an additional efficiency calibration has to be executed in order to determine the full-energy peak efficiency factor ε. The full-energy peak efficiency factor is defined as the fraction of photons of a certain energy emitted by a source that contributes to a full-energy peak observed in the pulse height spectrum (of the corresponding energy). [14] [41]

The efficiency depends on the geometry of a sample, the density and size of the sample and the distance between the detector and the source. Other factors that influence the full-energy peak efficiency factor are the photon energy and the detector specifications and setup. It is therefore only possible to perform an adequate efficiency calibration with a calibration source which has the same properties as the samples and has a sufficient range and number of energy peaks. [14] [41]

For the efficiency calibration, a multi-gamma source was placed in the same petri dish geometry as the samples. The composition of the used multi-gamma source and the corresponding energies and activities are illustrated in Table 4. The certification of the calibration source can be found in appendix 2.

Table 4: composition of the multi-gamma calibration source [44]

radionuclide	Energy (keV)	specific activity (kBq/g)	Extended relative uncertainty (k=2, %)
Am-241	59.5409 ± 0.0001	3.17 x 10^1	± 4
Cd-109	88.0336 ± 0.0001	2.78 x10^2	± 5
Co-57	122.06065 ± 0.00012	1.560 x10^1	± 3
	136.47356 ± 0.00029		
Ce-139	165.8575 ± 0.0011	1.645 x 10^1	± 4
Cr-51	320.0835 ± 0.0004	2.35x10^2	± 4.5
Sn-113	391.698 ± 0.003	4.62x10^1	± 4
Sr-85	514.0048 ± 0.0022	4.89 x 10^1	± 3.5
Cs-137	661.657 ± 0.003	6.08 x 10^1	± 3
Co-60	1173.228 ± 0.003	8.78 x 10^1	± 3
	1332.492 ± 0.004		
Yt-88	1836.052 ± 0.013	9.62 x 10^1	± 3

The calibration source was measured in direct contact with the HPGe-detector. Figure 28 shows the measured full-energy peak efficiency curve. The equation of the measured efficiency curve was of the form:

$$\ln(\varepsilon) = -2.198 * 10^2 + 1.893 * 10^2 * \ln(E) - 6.487 * 10^1 * \ln(E)^2 + 1.109 * 10^1 * \ln(E)^3 - 9.31 * 10^{-1} * \ln(E)^4 + 3.131 * 10^{-2} * \ln(E)^5$$

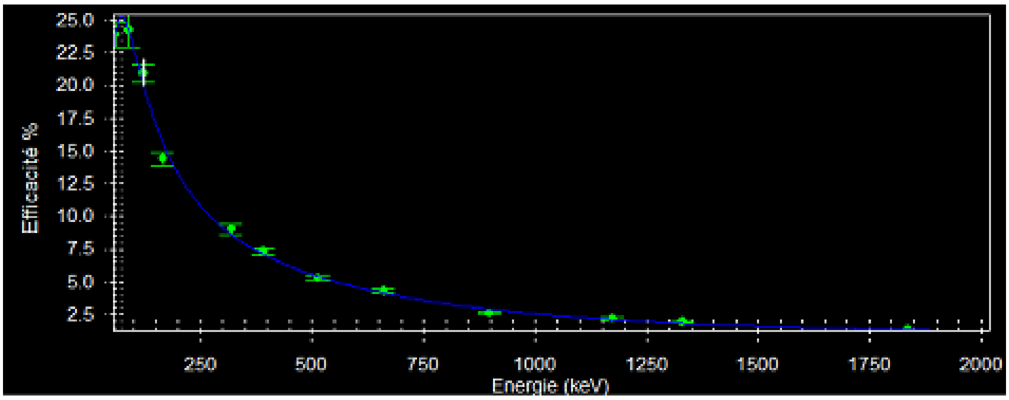

Figure 28: full-energy peak efficiency curve

Apex gamma software uses the efficiency equation in combination with the branching ratio, measuring time and the peak counts to calculate the activity of the sample. The specific activity can then be calculated by dividing the activity by the mass of the sample. [45]

For the measurement of the surface contamination, the efficiency curve was measured with a different source than the one listed above. The sources were placed in the middle of the detector and in direct contact to the surface of the detector. Since the geometry of the calibration source is different to the geometry of the sample, an error is to be expected for the calculated activity. Because the calibration source, as well as the measured swipe samples, has a planar geometry and they were both measured at the surface, the error is expected to be small. The technique of using swipe samples is considered to be inaccurate because of the big uncertainty on the pick-up factor. The error on the efficiency curve for the swipe samples is therefore considered to be of less importance than the error coming from the uncertainty of the pick-up factor. [43]

8.4.4 Output data of the HPGe-detector

As was mentioned previously, the data of the spectra were analysed with Apex Gamma software of Canberra Industries. The resulting analysis report consists of the following components: [45]

- A peak search report;
- A peak identification report;
- The results of the quantitative analysis.

The goal of the peak search report is to locate all relevant peaks which are located in the measured spectrum. The regions where the peaks are located are called regions of interest (ROI). The peak search report contains the found peaks with the corresponding energy and the start and end of the ROI. Furthermore the report also gives the channel number of the centre of the located peak, the surface area of the peak with the corresponding uncertainty, the background of the peak and the full with half maximum (FWHM) of the peak. [45]

The peak identification report appoints the correct nuclide to the found peaks of the peak search report. The identification of the nuclides is carried out with nuclide libraries that list radionuclides with their corresponding half-live times, energy lines and the yield of each line. The result of the comparison of the found peaks with the internal library is a list of identified nuclides in the spectrum. [45]

The last part of the analysis report gives the results for the quantitative analysis. The activity of the sample is calculated with the following factors: [45]

- The branching ratio of the identified nuclides;
- The full-energy peak efficiency factor ε of the corresponding energy;
- The net peak area;
- Time of the collection.

Extra corrections are also performed for the nuclide decay during counting. The specific activity of the sample can be obtained by dividing the calculated activity by the mass of the sample. Next to the activity of the sample, the total uncertainty of the activity is also displayed in the report. The random uncertainty of the activity (σc) is a composition of the following uncertainties: [45]

- The user defined random uncertainty;
- The uncertainty of the net peak area;
- The uncertainty of the sample quantity;
- The uncertainty of the efficiency calibration;
- The uncertainty of the activities of the calibration gamma source;
- The uncertainty of the branching ratio;
- The half-life uncertainty.

The total uncertainty (σc(T)), as defined in the report, can then be calculated with the random uncertainty of the activity (σc) and the user defined systematic uncertainty (σ_{sys}). The following formula is used for the calculation of the total uncertainty: [45]

$$\mathrm{\sigma c(T)} = \mathrm{\sigma c} + \frac{\mathrm{\sigma sys} * \mathrm{C}}{100}$$

In certain specific situations, the report did not identify all the peaks measured in the spectrum. A manual check of the spectrum was therefore always performed to avoid incorrect or lack of information. This was carried out by checking both the presence of the found peaks in the report of the spectrum and checking the spectrum for additional non identified peaks. For this control, the online spectrum catalogues of the Idaho national laboratory as well as the Lund/LBNL nuclear data search tool were used. [46] [47]

The activities of all the 75 samples that were taken varied widely. In order to obtain an adequate spectrum and uncertainty, different measurement times were applied. The measuring times are listed in the table below.

Table 5: measurement time of the samples

measurement time	number of samples
500 s	10
1000 s	12
1 h	26
10 h	24
24 h	3

9 Results and discussion

In total 75 samples were taken and measured. 59 swipe samples were taken in the hot cells and glove boxes. 7 swipe samples were taken of the target mount and targets. The resulting 16 samples were drill samples which were taken in the targets and the target mounts. The peaks that were used to identify the radionuclides in the samples can be found in in annex 3.

The results of the swipe samples are expressed in Bq/cm^2 and can be interpreted as a semi quantitative result of the non-fixed contamination. A swipe efficiency of 20% will be used, in accordance with the approved procedure for the collection of swipe samples, established by AV Controlatom. For the efficiency calibration of the swipe samples, general calibration sources were used. The use of a calibration source with the exact same geometry as the swipe samples would have been more correct but such a source was not present on the site. It was also not seen as a priority because the used calibration source is also planar and thus the efficiency will not deviate much in comparison with a source with the exact same geometry to the swipe samples. Furthermore, the uncertainty of the swipe sample efficiency will be far greater than the introduced error of the efficiency with the used source. [40] [22]

The results for the drill samples are expressed in Bq/g and can be interpreted as a quantitative result of the activation. The efficiency calibration was performed with a calibration source with the exact same geometry to the measuring geometry of the samples. The geometry that was used was already elaborated in chapter 8.

9.2 Swipe samples of the hot cells

In total, 40 swipe samples were taken in the hot cells. The samples were taken on the four walls of the hot cells (left wall, right wall, back wall and glass) and the work sheet of the hot cells.

When looking at the activities of the radionuclides in the hot cells, it can be noticed that the activity of the contamination on the work sheet is significantly higher than the activity on the side walls. This is an expected result because the irradiated targets were handled on top of this work sheet.

The results also show that the contamination is not only limited to the work sheet, but is also present on each side wall of the hot cell (although the activity is lower than for the work sheet). This is also an expected result because it was expected that the contamination would be present in the entire inner side of the alfa box of the hot cell.

Although several swipe samples were always taken in a same hot cell, not all of the swipe samples contained the same radionuclides. The explanation is that it is possible that the contamination which was sampled in the area of 300 cm^2 did not contain all the radionuclides present in the hot cell or that the quantity of the radionuclides is too small to be detected.

9.2.1 C30

The results of the swipe samples taken in hot cell C30 can be found in the table below.

Table 6: results for hot cell C30

C30	work sheet	right wall	left wall	glass wall	back wall
radionuclide 1	Na-22	Tl-202	Na-22	Na-22	Na-22
Act. (Bq/cm2)	6.3E-01	5E-02	2.4E-01	2.1E-01	2.03E-01
Unc. (Bq/cm2)	8E-02	2E-02	5E-02	6E-02	3.28E-02
radionuclide 2	Co-57				
Act. (Bq/cm2)	1.1E+00				
Unc. (Bq/cm2)	2E-01				
radionuclide 3	Co-60				
Act. (Bq/cm2)	3.4E-01				
Unc. (Bq/cm2)	4E-02				
radionuclide 4	Ag-108m				
Act. (Bq/cm2)	1.7E+00				
Unc. (Bq/cm2)	1E-01				
radionuclide 5	Tl-202				
Act. (Bq/cm2)	3.8E+00				
Unc. (Bq/cm2)	4E-01				
radionuclide 6	Tl-204				
Act. (Bq/cm2)	2.3E+02				
Unc. (Bq/cm2)	2E+01				
radionuclide 7	Bi-207				
Act. (Bq/cm2)	2.3E-01				
Unc. (Bq/cm2)	3E-02				

For hot cell C30, 7 different radionuclides were identified. These radionuclides are: Ag-108m, Na-22, Co-57, Co-60, Cd-109, Bi-207, Tl-204 and Tl-202. The presence of cobalt, thallium and silver radionuclides was unexpected in hot cell C30 because this cell was only used for the production of I-123. Further research into the use of this cell showed that in February 2005, a swipe sample was taken of the work sheet that showed the presence of thallium and cobalt radionuclides. The origin of this contamination is however not clear.

9.2.2 C31 chemistry

C31 chemistry was used to clean the irradiated targets, wash off and purify the irradiated targets. The results of the swipe samples can be found in the table on the next page. Hot cell C31 chemistry was used only for the production of Tl-201.

Table 7: results for hot cell C31 chemistry

C31 chem.	work sheet	right wall	left wall	glass wall	back wall
radionuclide 1	Cd-109	Co-57	Ag-108m	Co-57	Co-57
Act. (Bq/cm2)	1.4E+01	8E-02	3.3E+00	2.1E-01	1.9E-01
Unc. (Bq/cm2)	2E+00	2E-02	3E-01	9E-02	4E-02
radionuclide 2	Tl-202	Ag-108m	Cd-109	Cd-109	Ag-108m
Act. (Bq/cm2)	1.8E+00	1.6E-01	2.4E+01	1.5E+00	3.3E-01
Unc. (Bq/cm2)	2 E-01	2E-02	4E+00	2E-01	3E-02
radionuclide 3	Tl-204	Cd-109	Tl-204	Tl-202	Tl-202
Act. (Bq/cm2)	8.5E+02	6E-01	1.49E+03	6E-02	1.3E-01
Unc. (Bq/cm2)	4E+01	2E-01	8E+01	2E-02	3E-02
radionuclide 4		Tl-202		Tl-204	Tl-204
Act. (Bq/cm2)		5.4E-01		3.1E+01	5.3E-01
Unc. (Bq/cm2)		7E-02		2E+00	3E+00
radionuclide 5		Tl-204			Bi-207
Act. (Bq/cm2)		6.2E+01			8E-02
Unc. (Bq/cm2)		4E+00			2E-02

The radionuclides identified in hot cell C31 chemistry are: Ag-108m, Co-57, Cd-109, Bi-207, Tl-204 and Tl-202.

9.2.3 C31 distribution

C31 distribution was used as a connection between hot cell C32 and C31 chemistry. The results of C 31 distribution can be found in the table below.

Table 8: results for hot cell C31 distribution

C31 distr.	work sheet	right wall	left wall	glass wall	back wall
radionuclide 1	Ag-108m	Na-22	Na-22	Na-22	Ag-108m
Act. (Bq/cm2)	1.5E+00	5.0E-01	6.0E-01	6.9E-01	4.0E-01
Unc. (Bq/cm2)	1E-01	9E-02	7E-02	8E-02	6E-02
radionuclide 2	Cd-109	Cd-109	Ag-108m	Tl-202	Cd-109
Act. (Bq/cm2)	1.5E+01	1.1E+00	1.3E-01	1.1E+00	8E+00
Unc. (Bq/cm2)	2E+00	3E-01	1E-02	1E-01	2E+00
radionuclide 3	Tl-202	Tl-202	Tl-202	Tl-204	Tl-202
Act. (Bq/cm2)	4.1E+01	8.0E+00	1.1E+00	4.3E+01	2.9E+01
Unc. (Bq/cm2)	4E+00	8E-01	1E-01	2.5E+00	3E+00
radionuclide 4	Tl-204	Tl-204	Tl-204		Tl-204
Act. (Bq/cm2)	3.1E+03	3.1E+02	1.18E+02		1.69E+03
Unc. (Bq/cm2)	2E+02	2E+01	6E+00		9E+01
radionuclide 5	Bi-207				
Act. (Bq/cm2)	3.8E-01				
Unc. (Bq/cm2)	9E-02				

The radionuclides that were found in this hot cell are: Na-22, Ag-108m, Co-57, Cd-109, Bi-207, Tl-204 and Tl-202.

9.2.4 C32

Hot cell 32 was used to insert and remove of materials through its airlock and was used for in the production process of Tl-201 and Co-57. The radionuclides that were found in this hot cell are listed in the table below.

Table 9: results for hot cell C32

C32	work sheet	right wall	left wall	glass wall	back wall
radionuclide 1	Ag-108m	Co-57	Ag-108m	Co-57	Co-57
Act. (Bq/cm2)	1.2E+00	1.6E-01	1.5E-01	8E-02	1.2E-01
Unc. (Bq/cm2)	1E-01	5E-02	4E-02	3E-02	3E-02
radionuclide 2	Cd-109		Tl-202	Tl-202	
Act. (Bq/cm2)	1E+01		8.1E-01	5E-02	
Unc. (Bq/cm2)	1E+00		1E-01	3E-02	
radionuclide 3	Tl-204		Tl-204		
Act. (Bq/cm2)	1.79E+03		1.10E+02		
Unc. (Bq/cm2)	9E+01		7E+00		
radionuclide 4	Tl-202				
Act. (Bq/cm2)	2.3E+01				
Unc. (Bq/cm2)	2E+00				
radionuclide 5	Bi-207				
Act. (Bq/cm2)	2.1E-01				
Unc. (Bq/cm2)	5E-02				

Six radionuclides were identified in C32. These radionuclides are Ag-108m, Co-57, Cd-109, Bi-207, Tl-204 and Tl-202.

9.2.5 C33 distribution

Hot cell C33 distribution had the same function as C31 distribution in the production process. The table on the next page gives the results of the swipe samples in hot cell C33 distribution.

Table 10: results for hot cell C33 distribution

C33 distr.	work sheet	right wall	left wall	glass wall	back wall
radionuclide 1	Na-22	Co-57	Co-57	Co-57	Co-57
Act. (Bq/cm2)	1.26E+02	1.1E+00	1.5E+01	4.0E+01	4.0E+01
Unc. (Bq/cm2)	9E+00	2E-01	2E+00	6E+00	6E+00
radionuclide 2	Co-57	Co-60	Co-60	Ga-68	Ga-68
Act. (Bq/cm2)	9E+02	6E-01	5.1E-01	1.0E+02	1.1E+02
Unc. (Bq/cm2)	1E+02	1E-01	9E-02	2E+01	2E+01
radionuclide 3	Co-60		Ga-68	Bi-207	Bi-207
Act. (Bq/cm2)	7.9E+01		6E+01	1.0E-01	1.6E+00
Unc. (Bq/cm2)	5E+00		1E+01	5E-02	2E-01
radionuclide 4	Zn-65		Ag-108m		
Act. (Bq/cm2)	4.2E+01		1.4E-01		
Unc. (Bq/cm2)	5E+00		5E-02		
radionuclide 5	Ag-108m		Cd-109		
Act. (Bq/cm2)	1.11E+01		7E+00		
Unc. (Bq/cm2)	6E-01		1E+00		
radionuclide 6	Cd-109		Tl-202		
Act. (Bq/cm2)	3.0E+03		2.8E-01		
Unc. (Bq/cm2)	3E+02		7E-02		
radionuclide 7	Tl-204		Tl-204		
Act. (Bq/cm2)	5.4E+01		1.24E+02		
Unc. (Bq/cm2)	5E+00		9E+00		
radionuclide 8	Bi-207		Bi-207		
Act. (Bq/cm2)	8.6E+00		5.2E-01		
Unc. (Bq/cm2)	7E-01		9E-02		

The identified radionuclides for hot cell C33 distribution are Ag-108m, Na-22, Zn-65, Ge-68/Ga-68 Co-57, Co-60, Cd-109, Bi-207, Tl-204 and Tl-202.

9.2.6 C33 chemistry

C33 chemistry had the same function as hot cell C31 chemistry in the production process. The results of the measurements are listed in table 11. Hot cell C33 chemistry was used in the production of Tl-201, Ge-68 and Co-57.

Table 11: results for hot cell C33 chemistry

C33 chem.	work sheet	right wall	left wall	glass wall	back wall
radionuclide 1	Ag-108m	Co-57	Co-57	Ag-108m	Ag-108m
Act. (Bq/cm2)	1.1E-01	7E-02	8E-02	2.1E-01	2.6E-01
Unc. (Bq/cm2)	3E-02	3E-02	4E-02	2E-02	2E-02
radionuclide 2	Tl-202		Co-60	Cd-109	Tl-202
Act. (Bq/cm2)	5.8E-01		9E-02	9E-01	1.6E-01
Unc. (Bq/cm2)	8E-02		2E-02	3E-01	3E-02
radionuclide 3	Tl-204			Tl-202	Tl-204
Act. (Bq/cm2)	3.9E+01			5.5E-02	5.1E+01
Unc. (Bq/cm2)	3E+00			2E-02	3E+00
radionuclide 4				Tl-204	Bi-207
Act. (Bq/cm2)				1.7E+01	8E-02
Unc. (Bq/cm2)				3E+00	2E-02

The 7 identified radionuclides in C33 chemistry are: Ag-108m, Co-57, Co-60, Cd-109, Bi-207, Tl-204 and Tl-202.

9.2.7 C34

Hot cell 34 was used to insert and remove of materials through its airlock and was used for in the production process of Tl-201, Ge-68 and Co-57. The radionuclides that were found in this hot cell are listed in the table below.

Table 12: results for hot cell C35

C34	work sheet	right wall	left wall	glass wall	back wall
radionuclide 1	Co-57	Co-57	Co-57	Ag-108m	Co-57
Act. (Bq/cm2)	9E-01	2.9E-01	6.1E-02	2E-01	2.3E-01
Unc. (Bq/cm2)	2E-01	7E-02	2E-02	2E-01	5E-02
radionuclide 2	Ag-108m			Bi-207	Ag-108m
Act. (Bq/cm2)	1.9E-01			5E-02	9.2E-02
Unc. (Bq/cm2)	3E-02			2E-02	1E-02
radionuclide 3	Cd-109				Bi-207
Act. (Bq/cm2)	1.3E+00				4E-02
Unc. (Bq/cm2)	3E-01				2E-02

The radionuclides that were found in C34 are Ag-108m, Co-57, Cd-109 and Bi-207. The maximum activity of the radionuclides in C34 was (1.3 ± 0.3)E+00 Bq/cm^2 for Cd-109.

9.2.8 C35

Sorting cell C35 was used for the disassembly of the targets and their target mounts. The cell was used in the production of Tl-201, Ge-68 and Co-57. The results of the swipe samples taken in this cell are shown in the table below.

Table 13: results for hot cell C35

C35	work sheet	right wall	left wall	glass wall	back wall
radionuclide 1	Ag-108m	Ag-108m	Cd-109	Ag-108m	Ag-108m
Act. (Bq/cm2)	3.9E+01	1.03E+01	3.0E+01	5.2E-01	8.4E-01
Unc. (Bq/cm2)	5E+00	7E-01	4E+00	6E-02	8E-02
radionuclide 2	Co-57	Co-57	Tl-202	Tl-204	Co-57
Act. (Bq/cm2)	5.2E+03	1.2E+01	1.1E+00	3.2E+01	5.0E-01
Unc. (Bq/cm2)	8E+02	2E+00	4E-01	3E+00	1E-01
radionuclide 3	Co-60	Co-60	Tl-204		Tl-204
Act. (Bq/cm2)	2.0E+03	2.0E+00	1.37E+03		2.4E+01
Unc. (Bq/cm2)	1E+02	4E-01	7E+01		3E+00
radionuclide 4	Cd-109	Cd-109			Bi-207
Act. (Bq/cm2)	6.7E+02	3.3E+01			1.0E-01
Unc. (Bq/cm2)	9E+01	5E+00			6E-02
radionuclide 5	Bi-207	Bi-207			
Act. (Bq/cm2)	5.9E+01	1.9E+00			
Unc. (Bq/cm2)	7E+00	3E-01			
radionuclide 6	Zn-65	Tl-204			
Act. (Bq/cm2)	7.4E+02	4.4E+02			
Unc. (Bq/cm2)	9E+01	4E+01			
radionuclide 7	Na-22	Na-22			
Act. (Bq/cm2)	1.7E+01	9.2E-01			
Unc. (Bq/cm2)	5E+00	4E-01			

As can be seen in the table, hot cell C35 contains 9 radionuclides namely: Ag-108m, Co-57, Co-60, Cd-109, Bi-207, Zn-65, Na-22, Tl-204, Tl-202.

The activity of the radionuclides in the hot cells vary from a minimum of (5 ± 2)E-02 Bq/cm^2 to a maximum of (5.2 ± 0.8)E+03 Bq/cm^2. Table 14 gives the hot cells and the identified radionuclides.

Table 14: identified radionuclides for each hot cell

radionuclide	C35	C34	C33 dis.	C33 chem.	C32	C31 dis.	C31 chem.	C30
Ag-108m	x	x	x	x	x	x	x	x
Cd-109	x		x	x	x	x	x	
Tl-204	x		x	x	x	x	x	x
Tl-202	x		x	x	x	x	x	x
Bi-207	x	x	x	x	x	x	x	x
Na-22	x		x			x		x
Co-57	x	x	x	x	x	x	x	x
Co-60	x		x	x				x
Ge-68/Ga-68			x					
Zn-65	x		x					

Because hot cells C35 to C33 chemistry were used for the production of the same radionuclides, it can be assumed that all the identified radionuclides can be found in all of these glove hot cells. This assumption can also be made for hot cell C31 distribution and C31. These two hot cells were only used for the production of Tl-201.

9.3 Swipe samples of the glove boxes

The three glove boxes were used for the recycling of the target materials. The sampling was not performed with tele manipulator arms, but with the gloves attached to the glove box. Unlike the swipe samples in the hot cells, only 4 side walls of the glove boxes could be sampled. The back wall was not accessible without opening the glove boxes and risking a spread of the contamination. It was therefore opted to only sample the left wall, right wall, front wall and the work sheet of the glove boxes. In total, 12 swipe samples were taken in the glove boxes and measured with the HPGe detector.

In comparison with the swipe samples taken in the hot cells, it can be noticed that in general the activities of the swipe samples taken in the glove boxes are higher than the activities of the samples obtained in the hot cells. It can also be noticed that unlike for the hot cells, the spreading of the contamination is more equally distributed over the work sheet and the side walls of the glove box.

9.3.1 Glove box 1

The glove boxes were used to clean and prepare targets for the irradiation with the cyclotron. Glove box 1 was used to sand of the targets and prepare them for another layer of enriched Tl-203. The results of the swipe samples in glove box 1 can be found in the next table.

Table 15: results for glove box 1

Box 1	glass wall	work sheet	left wall	right wall
radionuclide 1	Ag-108m	Ag-108m	Ag-108m	Ag-108m
Act. (Bq/cm2)	2.4E+03	7.0E+02	2.3E+03	1.14E+03
Unc. (Bq/cm2)	1E+02	1E+01	1E+02	5E+01
radionuclide 2	Cd-109	Cd-109	Cd-109	Cd-109
Act. (Bq/cm2)	4.7E+03	7.0E+02	4.9E+03	2.6E+03
Unc. (Bq/cm2)	5E+02	7E+01	5E+02	3E+02
radionuclide 3	Tl-202	Tl-204	Tl-202	Tl-202
Act. (Bq/cm2)	8E+00	1.23E+03	1.1E+01	5E+00
Unc. (Bq/cm2)	5E+00	9E+01	5E+00	2E+00
radionuclide 4	Tl-204	Bi-207	Tl-204	Tl-204
Act. (Bq/cm2)	4.0E+03	3.3E+00	4.8E+02	1.9E+03
Unc. (Bq/cm2)	3E+02	9E-01	3E+01	1E+02
radionuclide 5	Bi-207		Bi-207	Bi-207
Act. (Bq/cm2)	3.6E+01		3.4E+01	1.6E+01
Unc. (Bq/cm2)	4E+00		4E+00	2E+00

The identified radionuclides present in the glove boxes are Ag-108m, Cd-109, Bi-207, Tl-204 and Tl-202.

9.3.2 Glove box 2

The second glove box was the electrodeposition station, and was used to electrodeposit a new layer of ^{203}Tl-enriched material on the cleaned (sanded) targets. The results of the swipe samples of glove box 2 are listed in the table below.

Table 16: results for glove box 2

Box 2	glass wall	work sheet	left wall	right wall
radionuclide 1	Ag-108m	Ag-108m	Ag-108m	Ag-108m
Act. (Bq/cm2)	5.5E+02	9.6E+02	6.4E+02	5.1E+02
Unc. (Bq/cm2)	3E+01	5E+01	3E+01	2E+01
radionuclide 2	Cd-109	Cd-109	Cd-109	Cd-109
Act. (Bq/cm2)	1.3E+03	1.7E+03	2.2E+03	9.8E+02
Unc. (Bq/cm2)	1E+02	2E+02	1E+02	1E+02
radionuclide 3	Tl-202	Tl-202	Tl-202	Tl-204
Act. (Bq/cm2)	6E+00	6E+00	5E+00	2.7E+03
Unc. (Bq/cm2)	1E+00	3E+00	2E+00	2E+02
radionuclide 4	Tl-204	Tl-204	Tl-204	Bi-207
Act. (Bq/cm2)	4.2E+03	1.62E+04	2.7E+03	7E+00
Unc. (Bq/cm2)	3E+02	9E+02	2E+02	1E+00
radionuclide 5	Bi-207	Bi-207	Bi-207	
Act. (Bq/cm2)	7.4E+00	1.4E+01	1.1E+01	
Unc. (Bq/cm2)	2E+00	3E+00	2E+00	
radionuclide 6		Na-22	Co-57	
Act. (Bq/cm2)		4E+00	9E+00	
Unc. (Bq/cm2)		1E+00	2E+00	

For glove box 2 the same radionuclides were found as in glove box 1. These radionuclides are: Ag-108m, Cd-109, Bi-207, Tl-204 and Tl-202. Next to these, two different radionuclides (Na-22 and Co-57) were also found.

9.3.3 Glove box 3

The third and last glove box is the assembly station. Here the electroplated targets were assembled with the target mounts and prepared to be re-irradiated. Table 17 shows the results of the swipe samples in glove box 3.

Table 17: results for glove box 3

Box 3	glass wall	work sheet	left wall	right wall
radionuclide 1	Ag-108m	Ag-108m	Ag-108m	Ag-108m
Act. (Bq/cm2)	3.7E+00	4.7E+01	2.7E+00	8.1E+00
Unc. (Bq/cm2)	2E-01	3E+00	2E-01	5E-01
radionuclide 2	Cd-109	Cd-109	Cd-109	Cd-109
Act. (Bq/cm2)	2.8E+01	1.3E+02	2.0E+01	2E+01
Unc. (Bq/cm2)	3E+00	1E+01	3E+00	3E+00
radionuclide 3	Tl-204	Tl-202	Tl-204	Tl-202
Act. (Bq/cm2)	2.3E+02	2.6E+00	4.7E+02	2E-01
Unc. (Bq/cm2)	1E+01	4E-01	3E+01	1E-01
radionuclide 4		Tl-204		Tl-204
Act. (Bq/cm2)		1.23E+04		1.21E+03
Unc. (Bq/cm2)		6E+02		6E+01
radionuclide 5		Bi-207		
Act. (Bq/cm2)		2.1E+00		
Unc. (Bq/cm2)		3E-01		

For glove box 3, the same radionuclides were found as in glove box 1. These radionuclides are Ag-108m, Cd-109, Bi-207, Tl-204 and Tl-202.

The activity of the radionuclides in the three glove boxes vary from a minimum of (2 ± 1)E-01 Bq/cm^2 to a maximum of (1.23 ± 0.06)E+04 Bq/cm^2. Although the 3 glove boxes were used in the same recycling process of the thallium targets, not all of the radionuclides were found in each box. Table 18 shows the found radionuclides in each glove box.

Table 18: identified radionuclides for each glove box

radionuclide	Box 1	Box 2	Box 3
Ag-108m	x	x	X
Cd-109	x	x	X
Tl-204	x	x	X
Tl-202	x	x	X
Bi-207	x	x	X
Na-22		x	
Co-57		x	

Because Na-22 and Co-57 were found in glove box 2, it is highly likely that these nuclides are also present in the other two glove boxes. It is therefore assumed that all the seven identified radionuclides can be found in all of the glove boxes.

9.4 Swipe samples of the activated materials

Next to the swipe samples in the hot cells and glove boxes, also swipe samples were taken of the activated materials. Targets 22 and 35 were used for the production of Co-57 and targets 1034 and 905 were used for the production of Tl-201. Next to these targets, two target mounts were also sampled (A11 and B14). The results of the swipe samples are shown in the table below

Table 19: results of the swipe samples of activated materials

	target 22	target 35	target 1034	target 905	mount A11	mount B14
radionuclide 1	Na-22	Na-22	Na-22	Na-22	Na-22	Na-22
Act. (Bq/unit)	1.4E+01	3.8E+01	4E+00	1.1E+01	3.6E+00	1.9E+01
Unc. (Bq/unit)	2E+00	6E+00	2E+00	2E+00	8E-01	2E+00
radionuclide 2	Co-57	Co-57	Co-57	Ag-108m	Co-57	Co-57
Act. (Bq/unit)	1.3E+02	1.3E+04	1.3E+01	6.2E+02	2.1E+01	2.7E+02
Unc. (Bq/unit)	2E+01	2E+03	3E+00	3E+01	3E+00	4E+01
radionuclide 3	Ag-108m	Co-60	Ag-108m	Cd-109	Ag-108m	Co-60
Act. (Bq/unit)	2.1E+01	5.21E+03	2E+03	4.1E+03	9.1E+01	9.4E+01
Unc. (Bq/unit)	1E+00	4E+02	1E+03	4E+02	5E+00	7E+00
radionuclide 4	Cd-109	Zn-65	Cd-109	Tl-202	Cd-109	Zn-65
Act. (Bq/unit)	5.1E+02	1.2E+03	1.1E+04	7+00	2.6E+02	1.9E+01
Unc. (Bq/unit)	5E+01	1E+02	1E+03	2E+00	3E+01	4E+00
radionuclide 5	Bi-207	Ga-68	Tl-204	Tl-204	Tl-204	Ga-68
Act. (Bq/unit)	3.3E+00	1.5E+04	1.4E+03	2.7E+03	2.9E+03	1.1E+02
Unc. (Bq/unit)	6E-01	2E+03	1E+02	2E+02	2E+02	5E+01
radionuclide 6	Co-60	Ag-108m	Bi-207	Bi-207	Bi-207	Ag-108m
Act. (Bq/unit)	3.5E+01	4.3E+01	1.2E+01	3.4E+01	2.2E+00	3.2E+01
Unc. (Bq/unit)	3E+00	4E+00	2E+00	3E+00	5E-01	2E+00
radionuclide 7		Cd-109				Cd-109
Act. (Bq/unit)		1.4E+03				1.2E+02
Unc. (Bq/unit)		1E+02				2E+01
radionuclide 8		Bi-207				Tl-204
Act. (Bq/unit)		8.3E+02				4.8E+02
Unc. (Bq/unit)		6E+01				4E+01
radionuclide 9						Bi-207
Act. (Bq/unit)						8E+00
Unc. (Bq/unit)						1E+00

Because the activated materials were stored on top of each other in the same hot cell, cross contamination of radionuclides coming from the production of Tl-201, Co-57 and Ge-68 is highly likely. The results of these swipe samples will therefore not be used to evaluate and link the measured radionuclides with the production process of its origin.

The results of these swipe samples will however be used to compare the radionuclides of the swipe samples with the radionuclides found in the drill samples of the activated materials. The goal of this comparison will be to see if there are additional radionuclides to be found in the bulk of the activated materials. The identified radionuclides on the swipe samples of the activated materials are: Na-22, Co-57, Co-60, Ag-108m, Cd-109, Bi-207, Tl-204, Tl-202, Zn-65 and Ga-68.

9.5 Drill samples of activated materials

In total, 16 drill samples were taken of activated materials. The activated materials that were sampled were the targets of both the Tl-201 and Co-57 production process and the target mounts.

Two Tl-201 targets (1034 and 905) were drilled on 3 different locations. The locations were the silver top layer, the copper layer and the weld. Target 905 was the most activated thallium target and number 1034 was the least activated. The most and least activated targets were chosen to investigate the difference between the two. For the two cobalt targets, the same sampling and measuring procedure was used as for the thallium targets.

Next to the targets, two mounts (A11 and B14) were also sampled. These mounts were sampled in the inox and the aluminium part.

It is expected that all the radionuclides that were found on the swipe samples can also be found in and on the activated materials. This is expected because the activated targets are the source of the contamination in the hot cells. Taking drill samples of the bulk of the activated materials was performed for two reasons. The first reason is to investigate if there are different radionuclides in the activated materials than in the contamination. The second reason is to calculate the mass specific activity in order to predict a final disposal option.

9.5.1 Tl-201 target 1034

Target number 1034 was a used for the production of thallium. Of the two sampled thallium targets, target 1034 was the least active with a contact dose rate of 737 µSv/h. The results of the drill samples for target 1034 can be found in the table below.

Table 20: results for target 1034

1034	copper	weld	silver
radionuclide 1	Co-60	Co-60	Na-22
Act. (Bq/g)	4.18E+02	9.7E+01	6E-01
Unc. (Bq/g)	7E+00	4E+00	3E-01
radionuclide 2	Ag-108m	Ag-108m	Ag-108m
Act. (Bq/g)	5.53E+02	1.09E+02	2.07E+03
Unc. (Bq/g)	8E+00	2E+00	3E+01
radionuclide 3	Cd-109		Cd-109
Act. (Bq/g)	2.7E+03		9.6E+03
Unc. (Bq/g)	1E+02		4E+02
radionuclide 4			Ag-110m
Act. (Bq/g)			5.0E+00
Unc. (Bq/g)			4E-01
radionuclide 5			Bi-207
Act. (Bq/g)			3.5E+00
Unc. (Bq/g)			5E-01

Six different radionuclides were identified in target 1034 with activities ranging from (6 ± 3)E-01 Bq/g to (9.6 ± 0.4)E+03 Bq/g. The identified radionuclides are Co-60, Cd-109, Ag-108m, Ag-110m, Na-22 and Bi-207. Of the identified radionuclides, no additional nuclides were identified to the ones observed in the swipe samples.

9.5.2 Tl-201 target 905

The second thallium target that was drilled was target number 905. This was the most activated thallium target with a contact dose rate of 1370 µSv/h. The identified radionuclides and their specific activities can be found in the table below.

Table 21: results for target 905

905	copper	weld	silver
radionuclide 1	Co-60	Co-60	Na-22
Act. (Bq/g)	6.47E+02	3.13E+02	3.9E+00
Unc. (Bq/g)	9E+00	5E+00	7E-01
radionuclide 2		Ag-108m	Ag-108m
Act. (Bq/g)		1.94E+02	5.35E+03
Unc. (Bq/g)		3E+00	7E+01
radionuclide 3		Ag-110m	Cd-109
Act. (Bq/g)		1.64E+01	4.3E+04
Unc. (Bq/g)		9E-01	2E+03
radionuclide 4			Ag-110m
Act. (Bq/g)			1.6E+01
Unc. (Bq/g)			1E+00

The identified radionuclides in target 905 are Co-60, Cd-109, Ag-108m, Na-22 and Ag-110m with specific activities from (3.9 ± 0.7)E+00 Bq/g to (4.34 ± 0.2)E+04 Bq/g. As with the previous target (1034), no additional radionuclides were found to the ones observed in the swipe samples.

When the results for target number 1034 (least activated) and 905 (most activated) are compared, it can be noticed that the specific activities of the radionuclides in target 905 are, as expected, higher that the specific activities of target 1034. The discussion to drill in the least and most activated target was also made in order to verify if different or more radionuclides are present in the most activated target. The identified radionuclides were however the same for both targets, with the exception of Bi-207 which was not found in target 905.

9.5.3 Co-57 target 22

The least activated Co-57 target (20 µSv/h) that was sampled was target 22. The results of the measurements can be found in the table on the next page.

Table 22: results for target 22

22	copper	weld	silver
radionuclide 1	Co-60	Co-57	Na-22
Act. (Bq/g)	6.9E+00	2.53E-04	1.73E+01
Unc. (Bq/g)	3E-01	4E-02	8E-01
radionuclide 2	Ag-108m	Co-60	Co-57
Act. (Bq/g)	2.5E-01	6.6E+00	1.5E+00
Unc. (Bq/g)	4E-02	2E-01	3E-01
radionuclide 3	Cd-109	Ag-108m	Ag-108m
Act. (Bq/g)	1.1E+00	7.9E+00	1.35E+03
Unc. (Bq/g)	4E-01	2E-01	2E+01
radionuclide 4		Cd-109	Cd-109
Act. (Bq/g)		2.5E+00	1.97E+04
Unc. (Bq/g)		6E-01	9E+02
radionuclide 5			Bi-207
Act. (Bq/g)			3.1E+00
Unc. (Bq/g)			8E-01

The radionuclides present in target 22 are Co-60, Co-57, Cd-109, Ag-108m, Na-22 and Bi-207. These nuclides were all found in the drill samples and had specific activities from (2.53 ± 0.04)E-04 Bq/g to (1.97 ± 0.09)E+04 Bq/g.

9.5.4 Co-57 target 35

The second cobalt target that was sampled was target 35. The contact dose rate of this target was 800 µSv/h. Table 23 shows the results of target 35.

Table 23: results for target 35

35	copper	weld	silver
radionuclide 1	Co-57	Co-57	Co-57
Act. (Bq/g)	5E-01	2.5E+00	6.8E+02
Unc. (Bq/g)	1E-01	3E-01	1E+01
radionuclide 2	Co-60	Co-60	Zn-65
Act. (Bq/g)	3.02E+02	1.40E+02	4.7E+01
Unc. (Bq/g)	4E+00	3E+00	8E+00
radionuclide 3		Ag-108m	Ag-108m
Act. (Bq/g)		4.42E+01	5.24E+03
Unc. (Bq/g)		9E-01	7E+01
radionuclide 4			Cd-109
Act. (Bq/g)			4.5E+04
Unc. (Bq/g)			2E+03
radionuclide 5			Bi-207
Act. (Bq/g)			1.8+01
Unc. (Bq/g)			1E+00

Six different radionuclides were identified. These radionuclides are Co-60, Co-57, Cd-109, Ag-108m, Zn-65 and Bi-207. The minimum specific activity was (5E ± 1)E-01 Bq/g, the highest specific activity was (4.5 ± 0.2) E+04 Bq/g.

The drill samples of the cobalt targets did not contain additional radionuclides than were found in the swipe samples. Although the Tl-201 and Co-57 used the same type of targets, no Ag-110m was found in the cobalt targets. The explanation for this is that the cobalt production was already abandoned in 2005 whereas the production of Tl-201 continued to 2012. Because of the shorter half-life of Ag-110m, it is likely that the Ag-110m in the cobalt targets has already decayed.

9.5.5 Target mounts A11 and B14

The last drill samples that were taken were samples of the aluminium and inox part of two target mounts. Mount A11 is the most activated and B14 is the least activated mount. The results of the drill samples can be found in the table below.

Table 24: results for target mount A11 and B14

	A11		B14	
	aluminium	inox	aluminium	inox
radionuclide 1	Mn-54	Mn-54	Co-57	Mn-54
Act. (Bq/g)	9E-01	2.95E+01	5.7E-01	5.3E+00
Unc. (Bq/g)	1E-01	9E-01	4E-02	3E-01
radionuclide 2	Co-60	Co-57	Co-60	Co-57
Act. (Bq/g)	2.2E+00	2.0E+00	7.4E-01	3.9E-02
Unc. (Bq/g)	1E-01	1E-01	8E-02	5E-03
radionuclide 3	Ag-108m	Co-60		Co-60
Act. (Bq/g)	1.9E-01	3.37E+02		7.1E+01
Unc. (Bq/g)	4E-02	5E+00		1E+00

The radionuclides identified in the mounts are Co-60, Co-57, Cd-109 and Mn-54. Mn-54 was not yet found in the swipe samples and can therefore be added to the list of found radionuclides.

9.6 Origin of the identified nuclides

By comparing the results of the glove boxes, hot cells and activated materials combined with literature, it is possible to evaluate and link the measured radionuclides with the production process of its origin.

By combining the results of the hot cells hot cells and glove boxes that were only used for the production of Tl-201, the origin of the identified radionuclides can be traced back to the Tl-201 production. The radionuclides that were found in C31 chemistry, C31 distribution and the three glove boxes are Tl-204, Tl-202, Na-22, Co-57, Zn-65, Ag-108m, Cd-109, Co-60 and Bi-207. Next to these radionuclides found in the hot cells and glove boxes, Ag-110m and Mn-54 are also added to the list of radionuclides coming from the production of Tl-201. Ag-110m is, just like Ag-108m, an activation product coming from the silver part of the target. This Ag-110m was found in the silver part of the drill samples taken in the target materials. Mn-54 was found in the target mounts that were used for both the production of Tl-201 and Co-57, and can thus be seen as a contaminant deriving from both the Co-57 and Tl-201 production process.

By comparing the results of the radionuclides identified as originating from the Tl-201 production with the radionuclides identified in the other hot cells, an estimation can be made to the origin of the other found radionuclides. The radionuclides that were found, and were not linked to the production of Tl-201, are Ga-68 and Co-60. Ga-68 derives from the production of Ge-68 and Co-60 is a side product of the production of Co-57. The list of radionuclides deriving from the production of Ge-68 also has to be extended with Co-57 because this radionuclide was an important side product of the Ge-68 production, as was already pointed out in chapter 6.4. The list of radionuclides coming from the Co-57 production process can also be complemented with the radionuclides found in the targets and the target mounts. Table 25 shows the link between the identified radionuclides and the production process of origin.

Table 25: origin of the identified radionuclides

origin	radionuclides	origin	radionuclides
I-123	/	Tl-201	Na-22
Ge-68	Co-57		Co-57
	Ge-68/Ga-68		Zn-65
Co-57	Na-22		Ag-108m
	Co-57		Ag-110m
	Zn-65		Cd-109
	Ag-108m		Mn-54
	Cd-109		Bi-207
	Mn-54		Co-60
	Bi-207		Tl-202
	Co-60		Tl-204

Combining the information from the literature regarding the function of the hot cells (figure 8) with the origin of the identified radionuclides (table 25), it is possible to predict the composition of the contamination in each hot cell and glove box. The table below shows the composition of the contamination that can be found in each hot cell and glove box.

Table 26: composition of the contamination of each hot cell and glove box

location	production process	Na-22	Co-57	Zn-65	Ag-108m	Ag-110m	Cd-109	Mn-54	Bi-207	Co-60	Tl-202	Tl-204	Ge-68 /Ga-68
C35	Tl-201, Co-57, Ge-68	x	x	x	x	x	x	x	x	x	x	x	x
C34	Tl-201, Co-57, Ge-68	x	x	x	x	x	x	x	x	x	x	x	x
C33	Tl-201, Co-57, Ge-68	x	x	x	x	x	x	x	x	x	x	x	x
C32	Tl-201, Co-57	x	x	x	x	x	x	x	x	x	x	x	
C31	Tl-201	x	x	x	x	x	x	x	x	x	x	x	
C30	I-123	x	x	x	x	x	x	x	x	x	x	x	
glove box 1	Co-57	x	x	x	x	x	x	x	x	x	x	x	
glove box 2	Co-57	x	x	x	x	x	x	x	x	x	x	x	
glove box 3	Co-57	x	x	x	x	x	x	x	x	x	x	x	

9.6.1 Germanium 68 and gallium 68

Germanium 68 is a radionuclide with a half-life of 271 days that decays through electron capture to gallium 68. The decay to gallium 68 has no gamma emissions, and thus Ge-68 cannot be measured with gamma spectroscopy. [48]

Germanium 68 was however indirectly measured through its daughter radionuclide gallium 68 which undergoes β^+-decay with a half-life of 68 days. The decay of Ga-68 results in the emission of photons and positrons. The Ga-68 that was measured in the samples originates from the production of Ge-68. Figure 29 shows the decay scheme of gallium 68. [46]

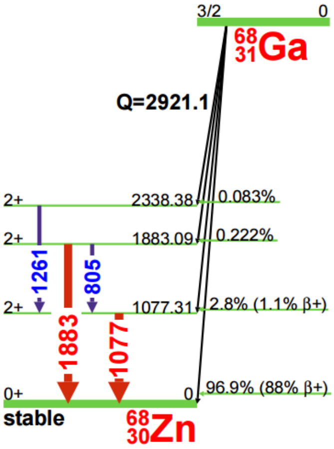

Figure 29: decay scheme of Ga-68 [46]

9.6.2 Cobalt 60 and cobalt 57

Cobalt 60 is a beta emitter with a half live of 5.27 years. Cobalt 60 is an activation product which can be formed in several nuclear reactions and is mostly formed as a result of the following reactions: [31] [48]

^{60}Ni (n, p) ^{60}Co	0.15769 barn (14 MeV neutrons)
^{63} Cu (n,α) ^{60}Co	0.037 barn (14 MeV neutrons)
^{59}Co (n, γ) ^{60}Co	37 barn (thermal neutrons)
^{61} Ni (n, np) ^{60}Co	0.015 barn (14 MeV neutrons)

The reaction with the highest cross section for the formation of Co-60 is the ^{59}Co (n, γ) ^{60}Co reaction with the 100% abundant stable isotope Co-59. Cobalt is a trace element that can be frequently found in carbon, steel and other alloys. Next to the neutron induced nuclear reactions, Co-60 was also produced as a side reaction to the Co-57 production trough proton induced reactions. [49] [30]

Cobalt 57 decays through electron capture and has a half-life of 271 days. This radionuclide was frequently found in the samples and originates from neutron and proton induced nuclear reactions. Co-57 was the product of interest in the Co-57 production and was formed with a (p,2n) or a (p, pn) reaction on Ni-58. Co-57 can also be produced trough (n, 3n) reaction of Co-59 and through (n, p + n) (p,2p) and (n, d) reactions of Ni-58. The decay schemes of Co-60 and Co-57 can be seen in the figure on the next page. [46] [48] [29]

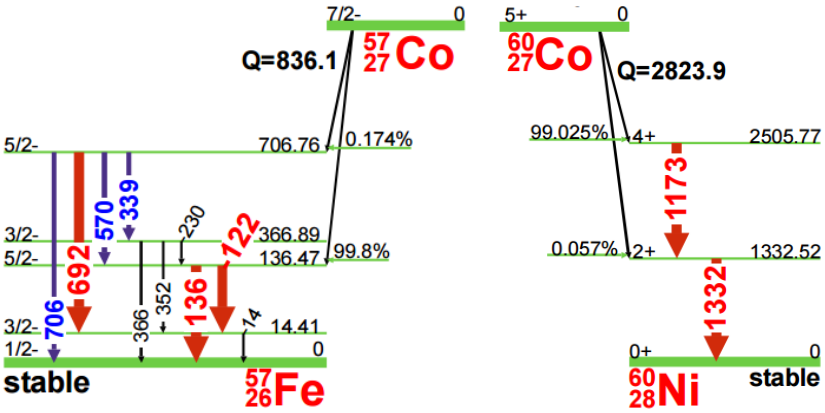

Figure 30: decay scheme of Co-57 and Co-60 [46]

9.6.3 Silver 108m and silver 110m

Both Ag-108m and Ag-110m are activation products that come from the activation of the silver layer of the targets. Silver 108m is produced by the ^{107}Ag(n, γ)^{108m}Ag reaction of the 51.8% abundant Ag-107. Ag-108m decays with a half-life of 130 years by electron capture to Ag-108 which decays to stable Cd-108 trough β^{-}-decay (2.4 minutes). [48] [49]

Silver 110m is a radionuclide that is produced with the ^{109}Ag(n, γ)^{110m}Ag reaction in the 48.2% abundant stable isotope. Ag-110m decays with a half-life of 249 days by β^{-}-emission to stable Cd-110. [48] [49]

9.6.4 Sodium 22

Sodium 22 is a radionuclide that is produced by nuclear reaction with fast neutrons on the 100% abundant Na-23. These reactions are the ^{23}Na(n, 2n)^{22}Na and ^{23}Na(γ, n)^{22}Na nuclear reaction. Na-22 decays by trough β^{+}-emission and gamma decay to stable Ne-22 with a half-life of 2.6 years. [48] [49]

9.6.5 Manganese 54

Manganese 54 is produced primarily by the ^{54}Fe(n, p)^{54}Mn nuclear reaction and decays through electron capture and a 835 keV gamma emission (312 days half-life). The target nucleus Fe-54 is 5.8% abundant and can be found in various alloys. [48] [49]

9.6.6 Bismuth 207

Bismuth 207 is produced by the irradiation of lead through a $^{207}Pb(p, n)^{207}Bi$ reaction. Bi-207 was observed multiple times and is believed to come from the welds on the target materials. Bi-207 is a radionuclide that decays by β^+-emission to stable lead 207 with a half-life of 31.5 years. [48] [49]

9.6.7 Zink 65

The $^{64}Zn(n, \gamma)^{65}Zn$ nuclear reaction of the 49% abundant Zn-64 is responsible for the production of Zn-65. The half-life of Zn-65 is 244 days, and Zn-65 decays via electron capture and by β^+-emission to stable Cu-65. Zinc is an element that is frequently found in variouse metal alloys. [48] [49]

9.6.8 Thallium 202 and thallium 204

Thallium 202 is a radionuclide that is a side product to the production of Tl-201. Tl-202 has a half-life of 12.23 days and decays by β^+-emission to Hg-202. It is produced with the following nuclear reaction: [27]

$$^{203}Tl\ (p,2n)\ ^{202m}Pb \text{ ---- (3.62 h) ---> } ^{202}Tl\ (12.23\ d)$$

Tl-204 is also a side product to the production of Tl-201. It is produced with the activation of Tl-203 by the $^{203}Tl(n, \gamma)^{204}Tl$ reaction. Tl-204 decays with electron capture and by β^--emission to respectively Hg-204 and Pb-204 with a half-life of 3.78 years. [27]

Its presence is visible during measurement by the X-ray analysis, but Tl-204 has the same x-rays than the Tl-202. Therefore, if Tl-202 is measured by its gamma rays, the presence of Tl-204 should always be suspected. In other words, if no Tl-202 is detected by its gamma rays, the X-ray peaks can be used to estimate the activity of Tl-204. If Tl-202 is present, the activity of the Tl-204 can be estimated with the ratio between the X-ray peaks and the height of the gamma peak at 439 keV. The excess of X-ray photons can then be linked to the presence of Tl-204. [48]

9.6.9 Cadmium 109

Cadmium 109 decays through electron capture to Ag-109 with a half-life of 461.9 days. Cd-109 can be formed on various ways. The main production modes of Cd-109 are: [31] [48] [50]

- $^{109}Ag\ (p, n)\ ^{109}Cd$;
- $^{110}Cd\ (p, pn)\ ^{109}Cd$;
- $^{108}Cd\ (n, \gamma)\ ^{109}Cd$.

10 Disposal options and future work

With the results of the activities of the activated materials and the contamination levels, it is now possible to evaluate the most suitable final disposal options. For the activated materials, the measured specific activities will be compared with the Belgian limits for unconditional release and the acceptance criteria of Studsvik and Energy*Solutions*. For the hot cells and glove boxes, a small preliminary study was made for the decontamination process.

10.1 Future of the activated materials

The Belgian limits for free release and the acceptance criteria of Studsvik and Energy*Solutions* were already discussed in chapter 7. A short overview of the conclusions is given in the table below.

Table 27: criteria for different disposal options

disposal option	criteria		
	dose rate (mSv/h)	specific activity for activated materials	specific activity for contaminated materials
unconditional release	/	$\sum_j Cj \,/\, Cj{,}L \leq 1$	$\sum_j Cj \,/\, Cj{,}L \leq 1$
melting: Studsvik	0.2 or 0.5	<1.2 kBq/kg for Co-60	<50 kBq/kg (β+ γ), <100 kBq/kg (α)
melting: Energy Solutions	0.2	1471 to 1375 kBq/kg	1471 to 1375 kBq/kg
cat. A radioactive waste	<5	above melting criteria	above melting criteria

For the target materials, the contact dose rate of target 1034, 905 and 35 exceeded the accepted dose rates for melting. Furthermore, the measured specific activities of all the targets were above the melting limits of both Studsvik and Energy*Solutions*. Only the dose rate of target 22 did not exceed 0.2 or 0.5 mSv/h but because the specific activity still exceeds the limits for melting, all the targets (and thus including target 22) can be classified as category A radioactive waste. Although only 11 targets were measured for their dose rates and only 4 of these 11 targets were drilled, it is expected that all of the target materials which are in hot cell C35 can be classified as category A waste.

For the target mounts, all the measured dose rates ranged between 1.1 and 6.3 µSv/h. The inox part of the mounts has a total specific activity of (3.69 ± 0.05)E+02 kBq/kg (for mount A11) and (7.6 ± 0.2) E+01 kBq/kg (for mount B14). As a result, the inox part of the targets can be melted at Energy*Solutions*. The aluminium part of the mount can be easily disassembled from the inox part and is less activated then the inox part. Although the aluminium part is less active, the specific activity is still above the limits for unconditional release.

10.2 Decontamination of hot cells and glove boxes

Choosing a decontamination technique suitable for the decontamination of the hot cells and glove boxes is a complex issue because the amount of decontamination techniques that are available is very diverse and large. The preliminary study for the decontamination of the hot cells was therefore made in consultation with experts of Tecnubel and the Institute for Radioelements (IRE). The flow chart below shows the strategy that can be used in the future to evaluate the future scenarios for the glove boxes and hot cells.

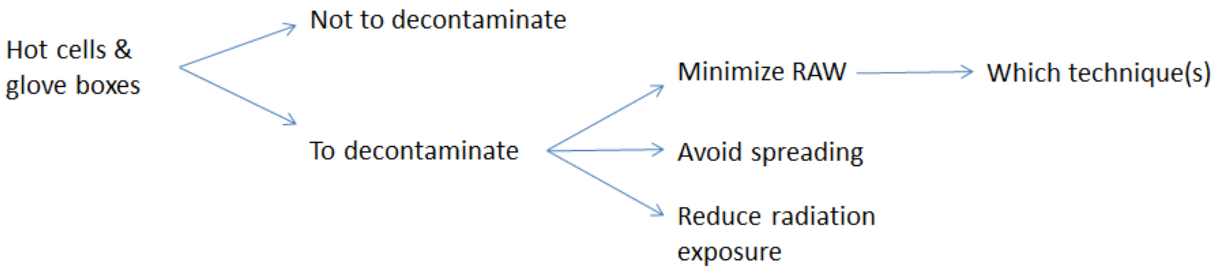

Figure 31: flow chart for the decontamination

The first step in the strategy is taking a decision to whether or not it is opportune to perform a decontamination on the hot cells and glove boxes. This choice depends on a number of factors such as technical, economical or safety reasons. Decontamination can be performed to achieve different goals. Making a choice to which goal that wants to be achieved is the second step in the flow chart. These goals can be the following: [51] [52]

- decontaminate in order to avoid the spreading of the contamination;
- decontaminate in order to reduce radiation exposure to workers;
- decontaminate in order to minimize the RAW.

Decontamination with the aim of minimizing RAW focusses on the removal of contamination as much as possible, in order to comply with the Belgian release limits and to liberate the previously contaminated materials. Decontaminations with the aim of releasing materials are much more extensive to normal decontaminations, and may require the use several techniques subsequent to each other. Making a decision about which technique or techniques will have to be used is the third step in the flow chart. The use of several techniques also means that these types of decontaminations are time consuming, and thus a compromise has to be made between the financial benefits of releasing the materials and the total cost of the decontamination. Another consideration that has to be made is the release limit. A first issue (see chapter 6) is that the limits are not expressed in Bq/cm^2 but in Bq/g. This problem can however be circumvented by dividing the total measured activity by the mass of the object. The second problem with the release limits is that not all of the measured radionuclides that were found in the contamination are included in the release limits. For example, Ga-68 and Ge-68 are not included in the annex IB of the ARBIS. [53] [18]

For the hot cells, the choice is already made to perform a decontamination. The initial goal will be to limit the amount of RAW by trying to release the inox alfa-box within the hot cells. This choice was made because the inox alfa-box of the hot cells is heavy and therefore expensive to dispose as radioactive waste. If the primary goal is not feasible with the decontamination process, a second objective can be decontaminate the alfa-box for melting at Studsvik or Energy*Solutions*. The approach for the decontamination was worked out in collaboration with Tecnubel and the IRE and the approach of the IRE was chosen for the decontamination. The procedure that is used by the IRE is however only intended to reduce the radiation doses in hot cells for maintenance work, and does therefore not focus on decontaminating to the extent of releasing. The quality of the decontamination can however be raised by repeating the decontamination procedure. Next to the chemical decontamination procedure of the IRE, it is expected that the chemical decontamination will have to be supplemented with a mechanical technique. The reason for the addition of the mechanical technique is that the chemical procedures which were used in the production of the radionuclides used strong acids. It is therefore believed that a contamination will in some places be more deeply into the surface of the alfa-box and thus more difficult to decontaminate. Furthermore, some spots of the hot cells show signs of corrosion which also makes it more difficult to decontaminate exclusively with chemical agents.

The chemical decontamination procedure consists of a succession of three chemical products. All the steps in the procedure are carried out with the telemanipulator arms of the hot cells. The chemical agents that are used in the procedure do not create additional waste or liquid waste. The first step of the decontamination is the removal of contaminated dust with a vacuum cleaner, followed by ALCATUM towelettes to remove any leftover dust and fine particles. The ALCATUM towelettes are pre-soaked towels soaked in ALCATUM, which is an alkaline mineral free decontamination solution. The next step of the decontamination uses an oxidizing gel named Fevdirad OX2 asp03. The gel needs to be applied to the desired surface and left to dry for two to three hours. Fevdirad OX2 is a gel that dries overtime and turns into a solid brittle layer that takes up radionuclides. Fevdirem BMD is then applied to the Fevdirad OX2 layer to weaken the dry film and neutralise the Fevdirad OX2. The resulting dried layer is shown in the figure below. [54]

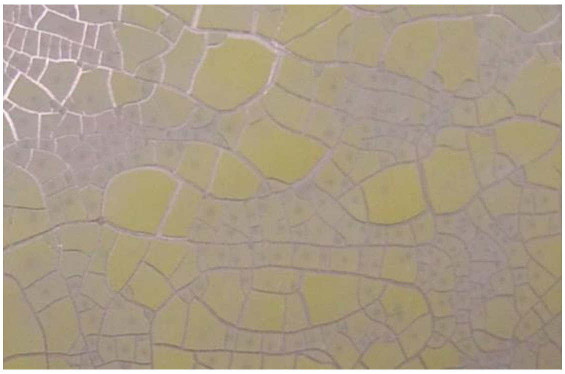

Figure 32: Fevdirad OX2 dry layer [54]

The resulting flakes can be removed with a vacuum cleaner. If there are still any hot spots left, for example on less accessible places such as corners and seams, a different type of gel can be applied on these locations (Fevdirad AC2 gel). This is an acid gel that does not congeal and is designed for the decontamination of stainless steel and lead. The final step in the chemical decontamination is the removal of the gel. If the contamination levels remain too high, the process can be repeated until the desired effect is achieved. [54]

For the mechanical decontamination technique, several options are available for decontaminating stainless steel components such as sand blasting, CO_2 blasting and several grinding and shaving techniques. For the chemical technique, not only the decontamination efficiency has to be taken into account, but also the availability of the equipment and expertise have to be considered. In consultation with experts on the site, the sponge jet decontamination technique was selected as one of the better candidates for the mechanical part of the decontamination. [51] [55]

Sponge blasting is an abrasive blasting techniques that uses a sponge media as abrasive medium. The sponge medium consists of a water based urethane sponge impregnated with abrasive materials. The abrasive materials can be steel grit, aluminium oxides, glass or other materials. The main reason for choosing sponge blasting over other blasting decontamination techniques is the limited amount of dust particles that is released during the blasting. In comparison with other blasting methods, the amount of dust that is set free during the process can be up to 99% less than with traditional abrasive blasting. The absence of dust is a major advantage because it prevents the spreading of radioactive contamination and also prevents the contamination of workers. The explanation for the limited amount of dust released lies within the different interaction mechanism of sponge media compared to conventional abrasive media. Figure 33 shows the interaction mechanism of conventional abrasive media compared to the interaction of sponge abrasive media. [51] [56]

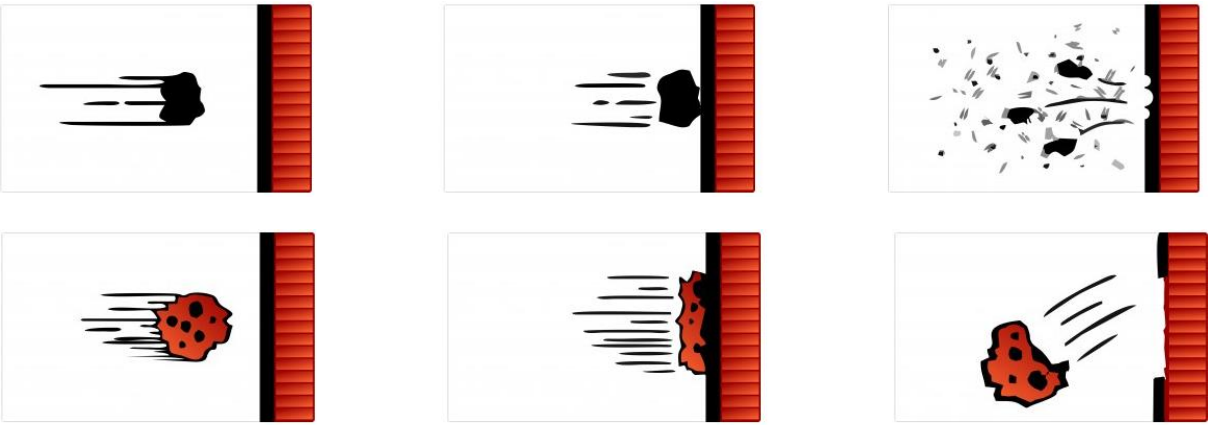

Figure 33: interaction mechanism of conventional abrasive media and sponge media [56]

The first three pictures show the interaction of a conventional abrasive media. The conventional media absorb the energy of the high speed collision by shattering and ricocheting into the air, creating airborne dust particles. With sponge media, the collision is absorbed through the flattening and deformation of the foam and the abrasives in the foam grate the surface. After the impact, the sponge media entrap most of the material that would have normally become airborne dust. A second advantage is that the sponge media can be reused up to ten times which reduces the secondary waste and disposal costs and also means that less abrasive material can be used. The media is recycled with a vibration classifier that separated the media from the waste fraction. [57] [58]

For the evaluation of the future disposal options of the glove boxes, the same flow chart can be used as for the hot cells. But unlike the hot cells, the glove boxes will however not be decontaminated for a number of reasons. The decontamination is not seen as useful because the glove boxes are made of plastic and have no reuse options after a possible decontamination, unlike metal from the hot cells that can be reused. Furthermore, because the glove boxes are made of plastic, they are more difficult to decontaminate and thus decontaminating to release levels would not be financially beneficial. The proposed final disposal scenario will therefore be as radioactive waste. Before going into the waste drums, the volume of the glove boxes will have to be reduced by cutting and dismantling the glove boxes in a glove tent. This is a tent that disposes of gloves, and can be placed over the existing glove boxes in order to retain the contamination when the glove boxes are decommissioned. Figure 34 shows dismantling operations with the aid of a glove tent. [59] [60]

Figure 34: glove box dismantling in a glove tent [61]

Before the hot cells and glove boxes can be dismantled or decontaminated, materials that are still present in the cells and boxes will have to be removed. These materials are contaminated and have a future disposal option as category A radioactive waste because decontaminating these components is not seen as financially beneficial. The materials that are still present in the hot cells are listed in appendix 5.

11 Conclusion

The focus of this master's thesis was to perform a radiological characterisation of glove boxes and hot cells present in the chemical zone of building B14 on the Fleurus nuclear site. This also included researching activated materials in the hot cells. The results of the measurements gave an identification of the radionuclides that can be found in the hot cells and glove boxes, as well as an indication of the contamination levels. Based on literature about the production of radionuclides, the origin of the identified radionuclides was also linked to the original production process.

In total 75 samples were sampled and characterised. The contamination in the hot cells was sampled with the tele manipulator arms of the hot cells and the sampling procedure was based on an approved method for taking swipe samples. In the hot cells and glove boxes, 59 swipe samples were taken and measured. For the activated materials, 7 swipe samples and 16 drill samples were characterized. In total, 13 radionuclides were found in the samples. These radionuclides are Na-22, Co-57, Co-60, Ag-108m, Ag-110m, Cd-109, Bi-207, Tl-204, Tl-202, Zn-65, Mn-54 and Ge-68/Ga-68.

The radionuclides that were linked to the cobalt production process are Na-22, Co-57, Co-60, Ag-108m, Cd-109, Bi-207, Zn-65 and Mn-54. These radionuclides were also linked to the thallium production with the addition of Ag-110m, Tl-202 and Tl-204. For the germanium production, only Co-57, Ga-68 and Ge-68 was found in the contamination.

Next to the identification of the radionuclides in the activated materials and contamination, the future disposal scenarios and decontamination options were further elaborated. For the hot cells, a decontamination approach was worked out with experts on the site. The proposed future approach for the decontamination starts with a chemical decontamination with ALCATUM, Fevdirad OX2 asp03 gel and Fevdirem BMD. After the chemical decontamination, a mechanical sponge blasting was chosen to follow up the chemical decontamination.

For the activated materials, the final disposal option will be as radioactive category A waste because both the specific activities as well as the contact dose rates are too high for either unconditional release or melting at specialized melting facilities. Next to the activated materials, other materials that are present in the hot cells will also be disposed as category A waste.

Finally, the future dismantling of the glove boxes was also discussed. The glove boxes will have to be dismantled in a glove tent before going into the waste drums as category A waste. The glove boxes will be dismantled in the glove tent in order to retain the contamination.

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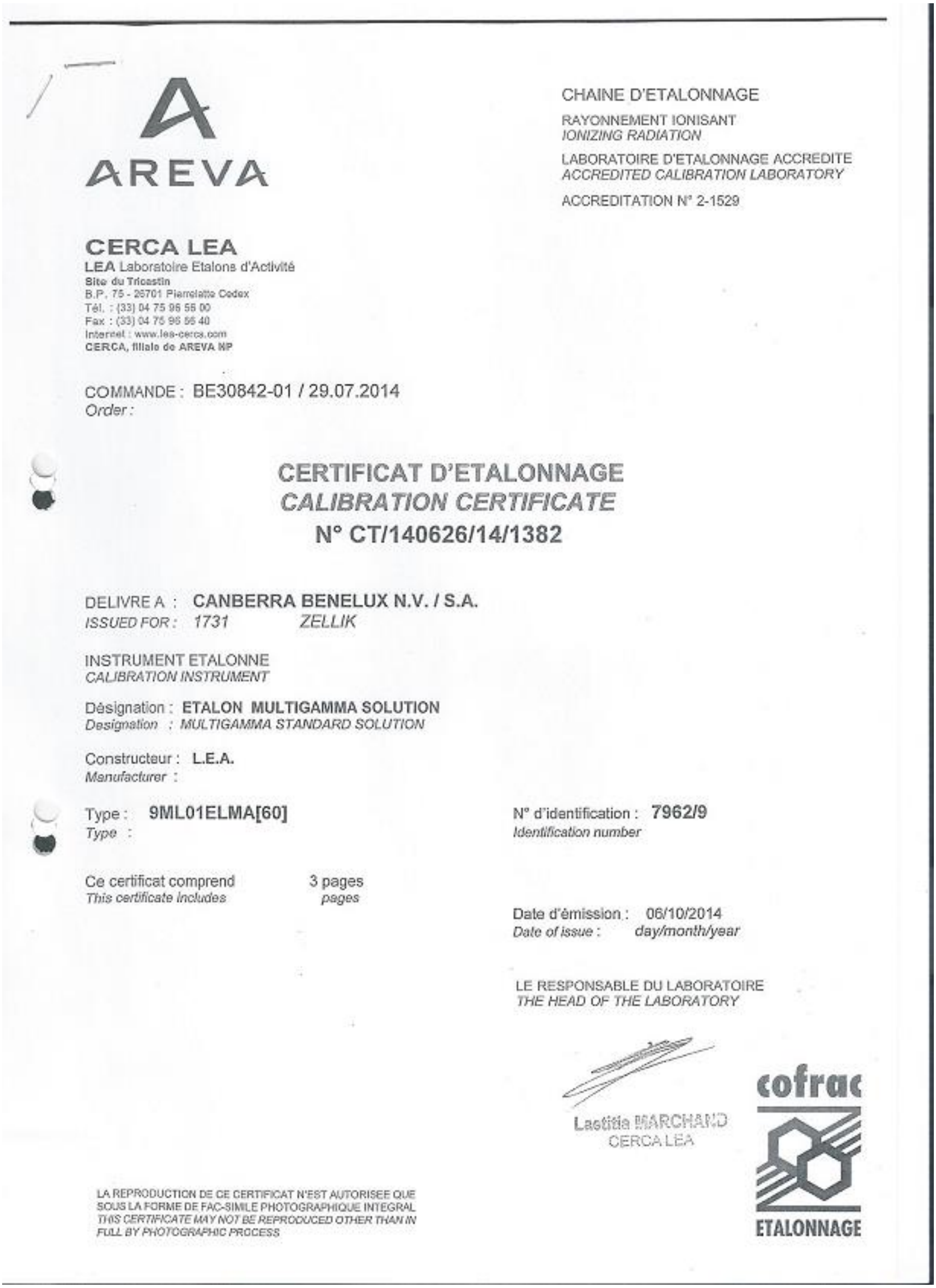
Appendix 1: dose rate measurements

production process	target number	front dose rate (µSv/h)	rear dose rate (µSv/h)
Tl-201	1134	750	205
	905	1370	429
	1018	1000	323
	1004	850	239
	1034	737	232
	941	1350	400
	1103	845	227
Co-57	21	392	81
	22	20	5
	10	121	30
	35	800	212
Tl-201, Co-57	A15	6,2	4,8
	A11	6,3	4,8
	A62	6	4,5
	B31	2,5	3,1
	A91	5,5	5,4
	A71	4,4	3,9
	B14	1,1	1,1

Appendix 2: calibration source



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Designation : MULTIGAMMA STANDARD SOLUTION

Constructeur : **L.E.A.**
Manufacturer :

Type : **9ML01ELMA[60]**
Type :

N° d'identification : **7962/9**
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1 Means and methods

Type of calibration	Photon flux
Unit	s^{-1}
Detector used	Solid state detectors Ge-HP (N)
Reference of the measurement equipment	CSGHP1/5
Method employed	γ-ray spectrometer

The environmental conditions have no influence on the results of the measurement.

2 Nominal characteristics delivered standards

Reference	Multigamma reference : **9ML01ELMA60**, N° **7962/9**
Type of container(*)	A
Daughter products	$^{113}In^{m}$, $^{109}Ag^{m}$, $^{137}Ba^{m}$
Volume	1 cm^{3}
Density	1,016 $g.cm^{-3}$
Chemical composition	Chloride of each component + $EuCl_3$ for ^{241}Am in HCl 1N
Reference date	**24/11/2014 at 12 h UTC**
Classification	No sealed source
No surface contamination (**)	Wipe test : **10/09/2014 OK**
Operator	E. ARMAING

(*) See product characteristics in LEA catalogue (www.lea-cerca.com).
(**) According to NF M61-003 / ISO 9978.

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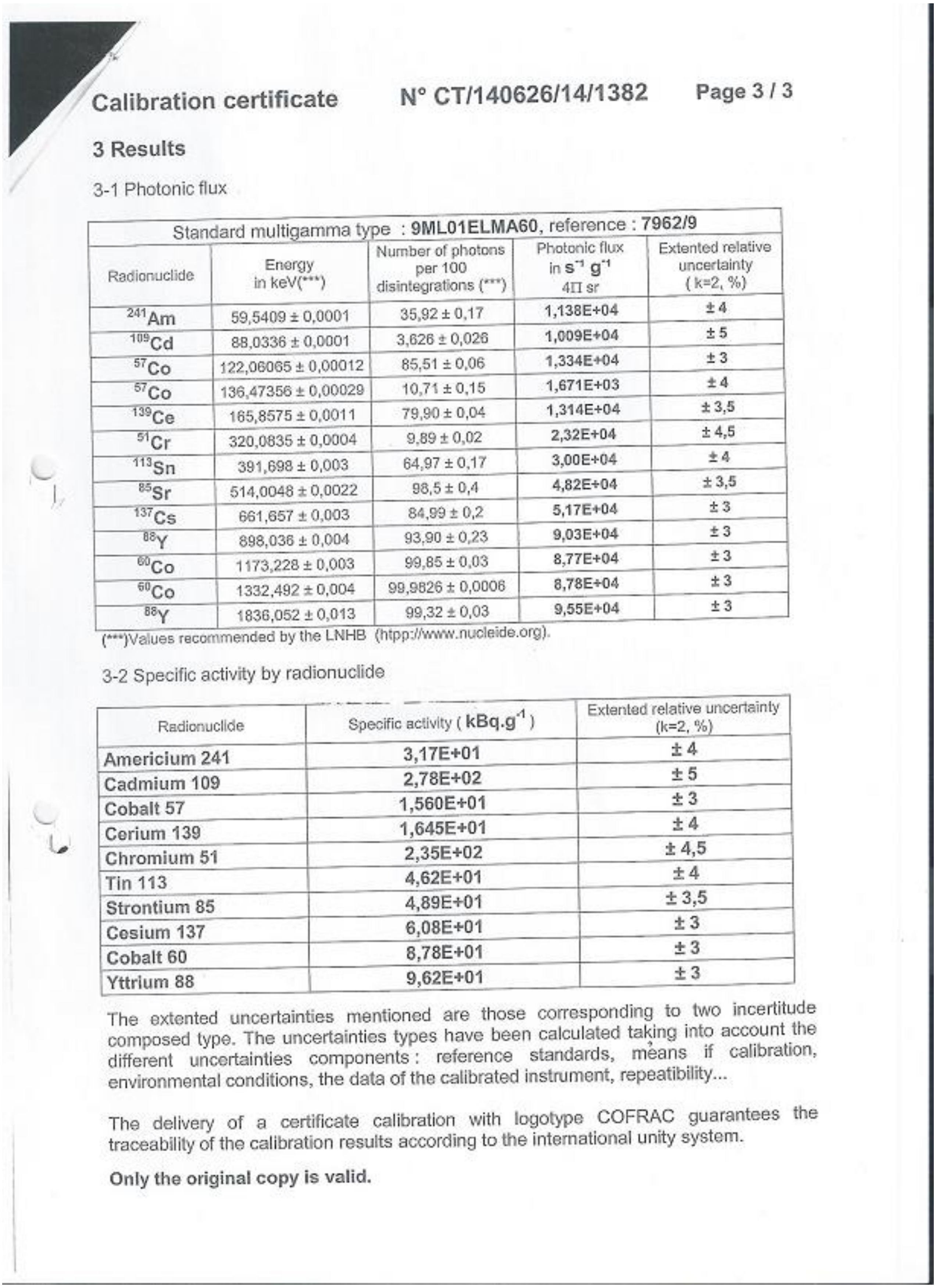

3 Results

3-1 Photonic flux

Standard multigamma type : **9ML01ELMA60**, reference : **7962/9**				
Radionuclide	Energy in keV(***)	Number of photons per 100 disintegrations (***)	Photonic flux in s^{-1} g^{-1} 4Π sr	Extented relative uncertainty (k=2, %)
^{241}Am	59,5409 ± 0,0001	35,92 ± 0,17	1,138E+04	± 4
^{109}Cd	88,0336 ± 0,0001	3,626 ± 0,026	1,009E+04	± 5
^{57}Co	122,06065 ± 0,00012	85,51 ± 0,06	1,334E+04	± 3
^{57}Co	136,47356 ± 0,00029	10,71 ± 0,15	1,671E+03	± 4
^{139}Ce	165,8575 ± 0,0011	79,90 ± 0,04	1,314E+04	± 3,5
^{51}Cr	320,0835 ± 0,0004	9,89 ± 0,02	2,32E+04	± 4,5
^{113}Sn	391,698 ± 0,003	64,97 ± 0,17	3,00E+04	± 4
^{85}Sr	514,0048 ± 0,0022	98,5 ± 0,4	4,82E+04	± 3,5
^{137}Cs	661,657 ± 0,003	84,99 ± 0,2	5,17E+04	± 3
^{88}Y	898,036 ± 0,004	93,90 ± 0,23	9,03E+04	± 3
^{60}Co	1173,228 ± 0,003	99,85 ± 0,03	8,77E+04	± 3
^{60}Co	1332,492 ± 0,004	99,9826 ± 0,0006	8,78E+04	± 3
^{88}Y	1836,052 ± 0,013	99,32 ± 0,03	9,55E+04	± 3

(***)Values recommended by the LNHB (htpp://www.nucleide.org).

3-2 Specific activity by radionuclide

Radionuclide	Specific activity (**kBq.g^{-1}**)	Extented relative uncertainty (k=2, %)
Americium 241	**3,17E+01**	**± 4**
Cadmium 109	**2,78E+02**	**± 5**
Cobalt 57	**1,560E+01**	**± 3**
Cerium 139	**1,645E+01**	**± 4**
Chromium 51	**2,35E+02**	**± 4,5**
Tin 113	**4,62E+01**	**± 4**
Strontium 85	**4,89E+01**	**± 3,5**
Cesium 137	**6,08E+01**	**± 3**
Cobalt 60	**8,78E+01**	**± 3**
Yttrium 88	**9,62E+01**	**± 3**

The extented uncertainties mentioned are those corresponding to two incertitude composed type. The uncertainties types have been calculated taking into account the different uncertainties components : reference standards, means if calibration, environmental conditions, the data of the calibrated instrument, repeatibility...

The delivery of a certificate calibration with logotype COFRAC guarantees the traceability of the calibration results according to the international unity system.

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Appendix 3: peaks of the identified radionuclides

radionuclide	half-life	energy (keV)	yield per decay (%)	decay type
Na-22	2,600 y	1274,54	99,94	β+
Co-57	270,900 d	122,06	85,51	EC
		136,47	10,6	
		692,03	0,157	
Co-60	5,271 y	1173,21	100	β-
		1332,48	100	
Zn-65	244,400 d	1115,52	50,75	β+
Ag-108m	418,000 y	79,14	6,63	β+, gamma
		433,94	90	
		614,28	89,8	
		722,91	90,8	
Cd-109	464,000 d	88,03	3,72	EC
Ag-110m	249,000 d	446,79	3,64	β-, gamma
		620,34	2,77	
		657,74	94,4	
		677,6	10,68	
		686098	6,47	
		706,67	16,68	
		744,26	4,64	
		763,92	22,28	
		818,01	7,3	
		884,66	72,6	
		937,47	34,2	
		1384,27	24,26	
		1475,75	3,97	
		1505	13,06	
		1562,26	1,18	
Bi-207	31,550 y	569,7	97,75	β+
		1063,66	74,5	
		1770,24	6,87	
Tl-202	12,230 d	439,56	91,4	β+
Ga-68	67,710 d	1077,35	3	β+
		1883,09	0,14	
Mn-57	312,700 y	834,82	99,975	EC

Appendix 4: release levels

Nuclide	liberation level (Bq/kg)
H-3	100
Be-7	10
C-14	10
Na-22	0.1
P-32	100
P-33	100
S-35	100
Cl-36	1
K-40	1
Ca-45	100
Ca-47	1
Sc-46	0.1
Sc-47	10
Sc-48	0.1
V-48	0.1
Cr-51	10
Mn-52	0.1
Mn-53	1000
Mn-54	0.1
Fe-55	100
Fe-59	0.1
Co-56	0.1
Co-57	1
Co-58	0.1
Co-60	0.1
Ni-59	100
Ni-63	100
Zn-65	1
Ge-71	10000
As-73	100
As-74	1
As-76	1
As-77	100
Se-75	1
Br-82	0.1
Rb-86	10
Sr-85	1
Sr-89	10
Sr-90+	1
Y-90	100
Y-91	10
Zr-93	10
Zr-95+	0.1
Nb-93m	100
Nb-94	0.1
Nb-95	1
Mo-93	10
Mo-99+	1
Tc-96	0.1
Tc-97	10
Tc-97m	10
Tc-99	1
Ru-97	1
Ru-103+	1
Ru-106+	1
Rh-105	10
Pd-103+	1000
Ag-105	1

Nuclide	liberation level (Bq/kg)
Ag-108m+	0.1
Ag-110m+	0.1
Ag-111	10
Cd-109+	10
Cd-115+	1
Cd-115m+	10
In-111	1
In-114m+	1
Sn-113+	1
Sn-125	1
Sb-122	1
Sb-124	0.1
Sb-125+	1
Te-123m	1
Te-125m	100
Te-127m+	10
Te-129m+	10
Te-131m+	0.1
Te-132+	0.1
Te-134	0.1
I-125	1
I-126	1
I-129	0.1
I-131	1
Cs-129	1
Cs-131	1000
Cs-132	1
Cs-134	0.1
Cs-135	10
Cs-136	0.1
Cs-137+	1
Ba-131	1
Ba-140	0.1
La-140	0.1
Ce-139	1
Ce-141	10
Ce-143	1
Ce-144+	10
Pr-143	100
Nd-147	10
Pm-147	100
Pm-149	100
Sm-151	100
Sm-153	10
Eu-152	0.1
Eu-154	0.1
Eu-155	10
Gd-153	10
Tb-160	0.1
Dy-166	10
Ho-166	10
Er-169	100
Tm-170	10
Tm-171	100
Yb-175	10
Lu-177	10
Hf-181	1
Ta-182	0.1

Nuclide	liberation level (Bq/kg)
W-181	10
W-185	100
Re-186	10
Os-185	1
Os-191	10
Os-193	10
Ir-190	0.1
Ir-192	0.1
Pt-191	1
Pt-193m	100
Au-198	1
Au-199	10
Hg-197	10
Hg-203	1
Tl-200	1
Tl-201	10
Tl-202	1
Tl-204	10
Pb-203	1
Pb-210+	0.01
Bi-206	0.1
Bi-207	0.1
Bi-210	10
Po-210	0.01
Ra-223+	1
Ra-224+	1
Ra-225	1
Ra-226+	0.01
Ra-228+	0.01
Ac-227+	0.01
Th-227	1
Th-228+	0.1
Th-229+	0.1
Th-230	0.1
Th-231	100
Th-232+	0.01
Th-234+	10
Pa-230	1
Pa-231	0.01
Pa-233	1
U-230+	1
U-231	10
U-232+	0.1
U-233	1
U-234	1
U-235+	1
U-236	1
U-237	10
U-238+	1
Np-237+	0.1
Np-239	1
Pu-236	0.1
Pu-237	10
Pu-238	0.1
Pu-239	0.1
Pu-240	0.1
Pu-241	1
Pu-242	0.1

Nuclide	liberation level (Bq/kg)
Pu-244+	0.1
Am-241	0.1
Am-242m+	0.1
Am-243+	0.1
Cm-242	1
Cm-243	0.1
Cm-244	0.1
Cm-245	0.1
Cm-246	0.1
Cm-247+	0.1
Cm-248	0.1
Bk-249	10
Cf-246	10
Cf-248	1
Cf-249	0.1
Cf-250	0.1
Cf-251	0.1
Cf-252	0.1
Cf-253	1
Cf-254	0.1
Es-253	1
Es-254+	0.1
Es-254m+	1

Appendix 5: inventory hot cells and glove boxes

C30		glove box 1	glove box 2	glove box 3
never used	ethanol bottle	sand blaster	bottles of Tl solution	gloves
	scale	target shielding	lead shielding blocks	measuring beaker
	cables	gloves	gloves	bottle of water
	lead shielding blocks		stand	scale
	bottle shielding blocks			calibration weights
	bottle holder			
	calibration weights			
	tele manipulator fixing tool			
	gas pump			
	seat			

C35	C34	C33		C32	C31	
targets	empty SAS	plastic support stands	cardboard box	waste bucket	bottles of water	tape
target supports		waste bucket	tele manipulator fixing tool	waste bag	waste bag	saw
waste bag			tape		sand blaster	support stands
shuttles			cables			stands
			plugs			

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Richting: **master in de industriële wetenschappen: nucleaire technologie-milieutechnologie-radiochemie**
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Voor akkoord,

Brabants, Lowie

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