

“Radioactive Waste : Classification, Management and Radiation Study for Long Term disposal facilities”



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Final Project Work
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Abstract

The following paper describe our team final project work on “**Radioactive Waste : Classification, Management and Radiation Study for Long Term disposal facilities**”. This project, how suggested in the title, deals with the characterization of HLW in order to study its behaviour inside a long term disposal facility (Radioactive waste containment canisters), with a particular eye on the spread of radiations through these containment to make some observation on the managerial and radioprotectional point of view.

In the first part there's a deep analysis over all the main types of radioactive waste which are produced/accumulated by humans. Is analysed their classification, their isotopic composition, and there are conclusions about their role in the radioactive waste disposal.

In the second part there's the explanation of what type of canisters, HLW compositions and waste matrices were taken in account for our numerical simulations.

In the third part there's the explanation of our *Montecarlo* simulations made with FLUKA : the assumptions we made, the model we adopted and the results we got.

In the fourth, and last, part, there are our conclusions about the results we got from the simulations, what are their consequences and also some observations from the Radioprotectional end Economical point of view.

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1 Radioactive Waste

Introduction

This first part of our project is a general overview about the radioactive waste sector, with a characterization of all these particular types of waste based on their classification, with an in depth analysis of their radionuclide isotopic composition differentiating them relying on their origin and management.

1.1 Radioactive Waste Classification

The first step in the Radioactive Waste Management lies on the classification of the radio-toxicity level of such materials, in order to enhance appropriate disposal strategies and to respect safety requirements fixed by IAEA, which expressly states that at every step the radioactive materials shall be characterized and classified following the indications given by the regulatory body. The first international standards about this topic started to be published in the 70's , and were continuously subjected to changes all over the years, to keep up with the modern researches and needs; In this paper we will refer to the last General Safety Guide published by IAEA in 2009. Here the waste are classified in different groups based on their different Activity content (or in some cases the activity concentration), and on their radionuclides Half-lives. All can be resumed in the following image:

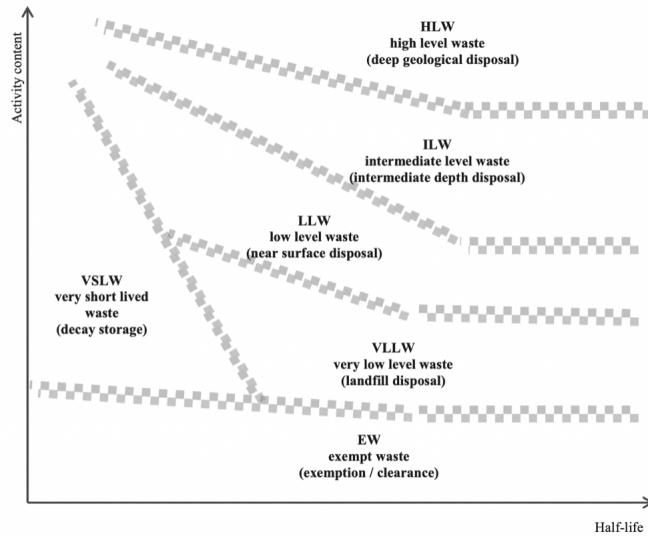


Figure 1: Illustration of the waste classification scheme (Ref.[1])

While the half-life is important to understand how the radiological hazard will evolve during the time, what type of containment is needed, and for how long the waste must be managed, the Activity content gives us indications of the need to contain such material, and to isolate it from the biosphere.

Although the IAEA gives the following classification, is important to keep in mind that these are general definitions and suggestions, more detailed quantitative boundaries to distinguish one class from the other are given directly by each state national program and requirement for nuclear disposal.

Looking closer to the waste characterization made by the IAEA, we can summarize in this way the main characteristics of each level individuated:

- **EXEMPT WASTE (EW):** The exempt waste category includes all

the materials constituted by a such low concentration of radionuclides, which is considered negligible. Once that a material is categorized as an exempt waste it doesn't need any type of further regulatory control in perspective (though they are not really treated as a regular waste from all points of view, for example there are limitations related to the discharge of these liquid/gaseous waste in the environment). For the maximum activity allowed to classify waste as exempts, see afterwards;

- **VERY SHORT LIVED WASTE (VSLW):** Very short lived waste are the ones which contains only very short half-life radionuclides over the clearance levels, and due to this they are stored until the activity level reaches values admitted for clearance, in order to treat them as traditional waste. As said the main criteria to classify them will be the half-life of the radionuclides, and also it's very important to determine the level of the long live radionuclides, which must be under acceptable levels. Usually the threshold fixed for the decay of the nuclides to reach clearance levels, is set to be around 100days or less;
- **VERY LOW LEVEL WASTE (VLLW):** Class of waste which comprehend all the materials that overcome the threshold of activity for the exempt waste, and that are not characterized by those very short half-lives of the VSLW. Usually they are disposed in surface landfill. There are no specific indications, in general the maximum activity to be considered VLLW is around 1 or 2 orders of magnitude above the exempt waste level (generally this is for artificial radionuclides, for the natural occurring ones is even lower due to the fact that they are characterized by longer half-lives activities);
- **LOW LEVEL WASTE (LLW):** In the previous classification kept until 2009 by IAEA the LLW were the ones that didn't require shielding during the normal applications and transports although their radioactivity, on the other hand the ones that required it (without needing also heat removal systems), were catalogued as ILW (Intermediate level waste), with the threshold between one and the other classification, fixed at 2mSv/h of contact radiation. With the new directives set by IAEA there's no more this such strong classification as the attention is prevalently given to long term safety. Since this class covers a very wide range of radionuclides, from the activity a little over the VLLW, up to waste which needs robust containment, there are no very specific international laws: every State has to give specific indications about the management of this class of waste, taking into account the specific radionuclides and the possibility of exposure; however some principle have to be considered, like the fact that periodical institutional controls must be guaranteed over a certain fixed time (usually around 300y), or the maximum limits fixed for the activity of long live alpha-emitting radionuclides in LLW (on average around 400Bq/g), and also for long live beta/gamma-radionuclides (up to tens of KBq/g). Generally these type of wastes are disposed in near surface facilities (note

that the classification of a waste in one category is also related to the selection of the disposal facility we want to do);

- **INTERMEDIATE LEVEL WASTE (ILW):** As seen above for the LLW, the distinction between this two categories is not very clear at the boundary, and it depends in large part in the selection made for the disposal facility we want to use. More in general the intermediate level waste are catalogued as waste that ‘contains long lived radionuclides in quantities that need a greater degree of containment and isolation from the biosphere than is provided by near surface disposal’ (Ref.[1]). The disposal of these waste has to be provided at depths between 10 and 100 meters in order to have no erosion of the containment, and have a better isolation from human possible contact with the waste;
- **HIGH LEVEL WASTE (HLW):** High level waste are all those waste of which radio-toxicity level is considered much higher than the ILW, needing a long term safety, provided by greater containment. One big difference to distinguish HLW from ILW is the fact that the first one generate non-negligible levels of decay heat during the centuries, and this must be eliminated. A typical concentration of activity of a HLW is in the range of $10^4/10^6 \text{ TBq}/m^3$ (the majority of these waste is generally nuclear spent fuel).

1.1.1 Exclusion, Exemption and Clearance Waste Classification

When someone is talking about radioactive waste, is very important to keep in mind these three words, and their meanings.

Treating radioactive sources, we are considering all the ones that are not subject to *exclusion* in the discussion. Excluded from the regulations are all the radioactive sources which are beyond human interference, such as the radionuclides present in our body (for ex. K40), the cosmic radiations, and so on. So when we will talk about radioactivity and dose, we will not consider the basic irradiation given by these sources, which basically are not amenable to control by humans.

After the elimination from the discussion of the ‘excluded materials’, another distinction must be done for the radio-toxic materials, dividing them in materials which are *exempt*, or not. With the term exempt, how said above in the classification, we consider all the materials (which on contrary of the excluded one, are amenable to be controlled) with radioactive characteristics, but such that they do not represent a healthy risk for the society, and due to this their treatment would be only a waste of society’s resources.

All the other radioactive waste are considered as relevant, and have to be treated until their radio-toxicity does no more exceed the levels of *clearance*. So cleared waste are basically the ones which had a relevant radio-toxicity, but that now, after an observation time under the regulatory control, are out of its jurisdiction. Here a simple image indicating the regulatory process for radioactive materials:

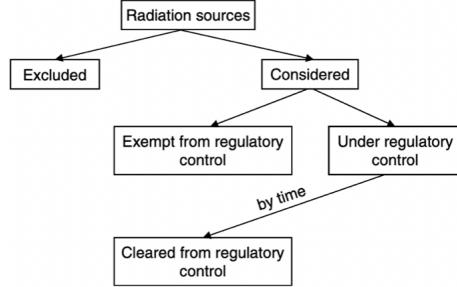


Figure 2: Regulatory Process for Radioactive Waste (Ref.[4])

The threshold set for the classification of the materials in the exempt category is (Ref.[4]):

- The effective dose is expected to be under 10microSv/y;
- Considering low probability scenarios of exposition, the effective dose must not be over the 1mSv/y;
- The Collective effective dose is not higher than 1 pers. Sv/y;

Other characteristics can be requested for particular radiation sources:

- Are automatically exempt all the radiation generators which in normal conditions do not cause an ambient dose equivalent rate of 1microSv/h at 0.1m of distance from any accessible surface, or that have a maximum energy generated of 5KeV;
- All the radionuclides from natural origin must be considered case by case, and they have a larger threshold, set to be 1mSv/y (higher due to the larger decay times that characterize the natural isotopes, and due to the presence of the natural background);

In the same way, are defined the clearance thresholds for the waste after a first period under the regulation controls (basically are considered cleared all the waste that, after time, enter into the levels of exemption listed above).

Note that to classify a certain material we go to watch the activity/activity concentration of the radionuclides into it, and we have then to compare them with the limits set , which must be respected in order to be considered exempt/cleared waste (see Tab.I1 (Ref.[4])).

While for the artificial waste characterized by mainly one radionuclide is sufficient to respect the activity levels listed in the tables, for materials characterized by a mixture of radionuclides, must be respected the following limit:

$$\sum_{i=1}^n \frac{C_i}{(\text{activity concentration})_i} \leq 1$$

Where C_i is the concentration of the i-th material, and the ‘activity concentration’ is the percentage of the i-th radionuclide activity given in respect to the

total one. The values to which refer are listed in specific tables, made by IAEA (Table 2, par.4, (Ref. [2])).

Starting from this moment we will refer Italian law decree to establish the thresholds of the nuclear waste (Ref.[5]) in a more precise and punctual way. Italy identifies three waste categories for what concern the classification with the half-life time :

- Very Short Life for those radionuclides with $T_{\frac{1}{2}} < 100d$;
- Short Life for those radionuclides with $100d < T_{\frac{1}{2}} < 31y$;
- Long Life with $T_{\frac{1}{2}} > 30y$.

Following then the indications cited above of IAEA, and also the ones from all the protection and surveillance association in the nuclear sector (like EURATOM, ICRP, ISIN, ...), are given more precise instructions dealing with the materials management with different type of activity in Italy' soil.

We have summarized all these things in the following table :

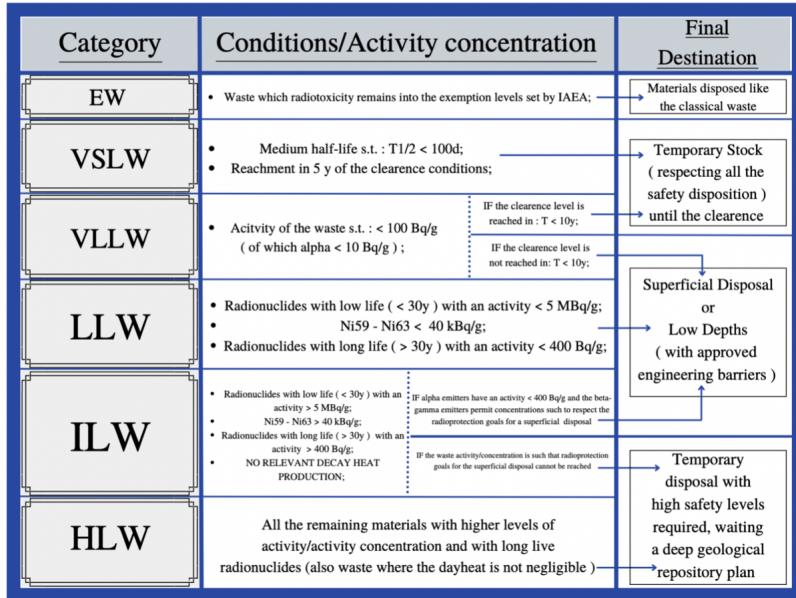


Figure 3: Italy Radioactive Waste Classification Scheme

1.2 NORM-TENORM Waste Characterization

In this chapter are both presented and characterized the NORM and TENORM waste.

With the term NORM are defined all the ‘Naturally Occurring Radioactive Materials’, which in essence are all the materials on the earth which contain, even in low, and different amount, primordial natural radionuclides such as Radium, Uranium, Thorium, Potassium, and their radioactive decay products (their characteristics will be seen below.).

Even if these types of materials could have inside a potential high level of radio toxic elements, usually in normal conditions the radiation spread by them is not worrying, and catalogued by ICRP into the every day background radiation.

Thus, their characterization remains very important, specially due to the fact that a lot of industrial activities come into contact with them, changing the isotopic concentration of them and/or exposing them to accessible environments, creating in this way what we use to call TENORM waste, which means ‘Technologically Enhanced NORM’;

These TENORM waste can be both formed by high quantities of low radioactive waste, or by lower concentration of materials with higher radio toxicity, depending on what type of technological operation they were involved in.

(Certainly they represent a non-negligible quantity of radioactive materials that has not to be ignored in our study on the Radioactive Waste Characterization.).

1.2.1 Radionuclides Characterization

Typically the primordial radionuclides cited are long-lived ones, characterized by half-lives in order of tens-, hundreds-, or even more, millions of years. They can be divided in two types of primordial radionuclides, the **non-series** ones, and the **series** ones, depending on if they are belonging to a particular decay pathway or not.

The non-series are the ones which not belong to any decay pathway and can be found singly.

Radionuclide	Half-life, y	Major Radiations	Typical Crustal Concentration, Bq/kg
⁴⁰ K	1.28×10^9	β, γ	630
⁵⁰ V	1.4×10^{17}	γ	2×10^{-5}
⁸⁷ Rb	4.75×10^{10}	β	70
¹¹³ Cd	9×10^{15}	β	$<2 \times 10^{-6}$
¹¹⁵ In	6×10^{14}	β	2×10^{-5}
¹²³ Te	1.24×10^{13}	x rays	2×10^{-7}
¹³⁸ La	1.05×10^{11}	β, γ	2×10^{-2}
¹⁴² Ce	$>5 \times 10^{16}$	β	$<1 \times 10^{-5}$
¹⁴⁴ Nd	2.29×10^{15}	α	3×10^{-4}
¹⁴⁷ Sm	1.06×10^{11}	α	0.7
¹⁵² Gd	1.08×10^{14}	α	7×10^{-6}
¹⁷⁴ Hf	2.0×10^{15}	α	2×10^{-7}
¹⁷⁶ Lu	3.73×10^{10}	β, γ	0.04
¹⁸⁷ Re	4.3×10^{10}	β	1×10^{-3}
¹⁹⁰ Pt	6.5×10^{11}	α	7×10^{-8}

Figure 4: Non-Series Radionuclides (Ref.[8])

As can be seen in the table above, while all the others concentrations are effectively negligible, K40 and Rb87 ones, are not. More in detail:

- **POTASSIUM** : In the three natural occurring potassium isotopes, only K40 is the one unstable, existing as an approximative constant of 0.0117% of stable potassium (mass ratio), with an emission of both β -rays (89%) with a maximum energy of 1,31MeV, and γ -rays (11%) with an energy of 1,46MeV. Due to its natural abundance, and to its high energy beta-particle emission, is the predominant radioactive component in humans every day life;
- **RUBIDIUM** : Rb87 exists with an atomic natural abundance on stable rubidium of 27.9%, with an emission of β -rays with a maximum energy of 0.275MeV. This primordial β -emitting radionuclide is present in the environment only at low concentrations.

Then there are the series one, which are all the isotopes belonging to the three main parent radionuclide decay pathway, that are U238, Th232, and U235, whose decay series are commonly called Uranium, Thorium and Actinium series respectively.

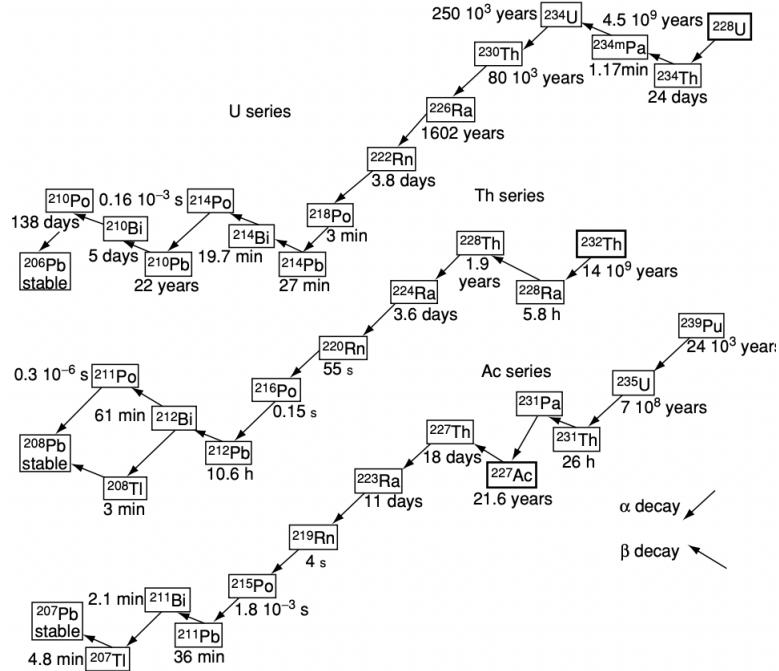


Figure 5: Visualization of the decay pathways pf 'series' primordial radionuclides (Ref.[7])

In the following lines will be described the most important radionuclides, which means that will be listed the ones that due to a scientific evidence result to be, either because of their concentration, either for their high level of radio-toxicity, the major contributors for human exposure to ionizing radiations.

- **URANIUM :** The primordial uranium that can be found in nature is formed almost completely by U238, which represents the 99.27% of the uranium mass, with the remaining part formed almost exclusively by U235, with a percentage of 0.72%, and by U234, with a percentage of 0.006% (for the study the remaining U's isotopes are negligible due to their such low presence in nature [U236, U233, U232]). Whether the U234 has a $T_{\frac{1}{2}} = 2,5 \times 10^5 y$, the both remaining have respectively an half life of $T_{\frac{1}{2}} = 7,1 \times 10^8 y$ (235), and $4,5 \times 10^9 y$ (238).
Uranium can be found in practically all the rocks and soils in different concentration, variating its presence from 0.5 ppm up to 4.7, corresponding to very different levels of activity between one rock and another.
Watching to the activity level of 1g of Natural Uranium, and how this is divided for its components : 0,33 μCi of 238, 0,33 μCi of 234 and 0,015 μCi of 235; this actually means that in normal conditions to obtain 1Ci of level of activity we would have to group 3 tonnes of natural uranium, which means that in nature is more relevant the damage given by the toxicity of this material than the radio toxicity of it. The things changes

when is considered enriched uranium, where the presence of U235 makes the material more dangerous from the point of view of the radio toxicity;

- **RADIUM** : Radium isotopes are extremely radioactive, and could be considered mainly 2 isotopes in nature : Ra226 and Ra228.
Ra 226 is a member of the decay path of U238, and it is one, if not the major, responsible to the fraction of the internal dose which is every year received by people from nature. It is characterized by a half life of 1600y, and it is an α emitter. To make a quick comparison with what said above for U, being the activity of Radium226 of 1Ci, the activity of 1g of it, it's equivalent to the activity of 3000kg of natural uranium. In addition to this Radium is not only a danger for its high level of radio-toxicity, but also for the danger that its progeny represents; Ra226 in fact has in its decay path Rn222, Pb210, Po210 and Bi 210, and so on... all dangerous radionuclides with high radio toxic levels. Like the uranium, also the radium226 is found in every rock and soil in variable amounts, and we can also refer to the presence of radium226 in them like the presence of U238 , material with which it is in roughly equilibrium. In addition to this, Radium has the tendency to precipitate with many minerals, thing that leads it to enter in water and in the natural alimentary human chain. Ra228 , differently from Ra226, is part of the decay chain of Th232. Although the occurrence in soil and water of Ra 226 and 228 is estimated to be in a 1:1 ratio, the radioactivity relevance of this element is not comparable to the one of Ra226. From the geochemical point of view is very similar to the Ra226, but their atomic properties and progenies are very different one from the other (the half life of Ra228 is $\tilde{5.75}$ y);
- **THORIUM** : The only primordial isotope of thorium findable in nature is Th232, but also shorter lived isotopes are present in all three natural decay chains listed above. Its natural composition is strictly related to the originally mineral, unlike Uranium. Its presence in rocks and soil is listed in the table below; its half life is $\tilde{1,4 \times 10^1}$ 0y (77000y for Th230, and 1.9y for Th228), and it has a high level radio toxicity, roughly 4.1kBq/g for Th232, and very similar for each daughter (Ref[5]). Due to its low mobility (exception made for ambient with a low-pH situation) Th232 is not present in biological elements in considerable amounts;
- **RADON** : There are three types of different isotopes of radon in nature, each one formed in a different decay path : Rn219 is part of the U235 chain, and it's the one which decays most rapidly, having a half life of roughly 3.96s (historical name: Actinon); Rn220 is a member of the Th232 chain, and it has a longer, but anyway short, half life than Rn219, which is 55.6s (historical name: Thoron); Rn222 is the most interesting Rn isotope in terms of radioactivity study if compared with the others two, and this is lead from the fact that it has a greater half life, roughly about 3,82d, it is part of the U238 chain, and is the element to which people classical refer when talking about 'radon'.

Radon has the characteristic to be a noble, non polar and inert gas. Usually Radon is trapped during the decay of its radium parent into rocks or soil, due to its gaseous form, has the tendency to recover into geologic fluids, mostly in water and soil gas, and then to reach the atmosphere when the soil gas enters in contact with the free surface. The radio toxicity of radon is really high, and it is very dangerous also because the radioactive decay products formed by Rn222 (and Rn220) are electrically charged, and tend to attach themselves to dust normally present in atmosphere, easy to be inhaled; so when this happen, all the Radon intake will decay until the achievement of a stabler isotopic condition: the 210Pb, which in human life span is a stable nuclide having a half life 22.3y. It's important to add that when 222Rn reaches the short term equilibrium, which means the 210Pb configuration, the energy released by the decays in human bodies is nearly 500 times greater than the only one derived from 222Rn itself.

- **LEAD and POLONIUM** : Pb210 is a member of the Rn222 chain (so of the U238 one), it's a beta emitter separated to its predecessor radon by six short live radionuclide decay (the longest radionuclide half-life between these two is the Pb214 with 26.8min), and that like suggested above, is considered with its 22.3y of half life a sort of 'stable' element. Pb210 will then eventually decay into the Po210 with a half life of 138.4d, passing through the Bi210, with a 5d half life. So as we could easily understand looking to all this half life data, after the decay of Rn222 in the atmosphere, Pb210 is produced rapidly, but due to its long life, he will decay only in the earth surface, coming down with precipitation (rain or snow).

In addition to all the previous radionuclides, which are either primordial ones, or part of their decay pathway, exist also other natural occurring radionuclides, which are the ones produced by the interaction between the cosmic rays and the atmospheric nuclei. From scientific observations we can say that the two most important of these radionuclides are **TRITIUM H3**, and **CARBON C14**, but that if compared with the primordial radionuclides, or with their progeny, in nature are not comparable in terms of radiation exposure. Instead, they will be considerate and characterized in the chapter related to the nuclear energy/research radioactive waste characterization.

Obs. Were discussed and characterized the radionuclides with the most relevant impact in term of : quality of the radiation (radio toxicity of the element itself), concentration on earth of the material, half life of the material, ...

Here are following the table cited for the radionuclides characterization.

Nuclide	Half-life	Source	Typical natural activity (Bq/kg)
¹⁴ C	5730 years	Cosmic ray interactions, ¹⁴ N(n, p) ¹⁴ C	220 in organic materials
³ H	12.3 years	Cosmic ray interactions with N and O; spallation from cosmic rays, ⁶ Li(n, α) ³ H	0.0012

Figure 6: Cosmogenic Radionuclides Chart (Ref.[7])

Material	^{40}K		^{232}Th		^{238}U	
	% total K	Bq/kg	ppm	Bq/kg	ppm	Bq/kg
<i>Igneous rocks</i>						
Basalt (crustal)	0.8	300	3-4	10-15	0.5-1	7-10
	1.1	300	2.7	10	0.9	10
Mafic						
Salic	4.5	1400	20	80	4.7	60
Granite(crustal)	>4.5	>1000	17	70	3	40
<i>Sedimentary rock</i>						
Shale	2.7	800	12	50	3.7	40
Sandstones						
	<1	<300	<2	<8	<1	<10
Clean quartz			2?	400?	3-6?	10-25?
Dirty quartz			2-3	600-900	2?	<8
					1-2?	10-25?
Arkose						
Beach sands	<1	<300	6	25	3	40
Carbonate rocks	0.3	70	2	8	2	25
All rock ^a	0.3-4.5	70-1400	2-20	7-80	0.5-4.7	7-60
Continental crust	2.8	850	10.7	44	2.8	36
Soil	1.5	400	9	37	1.8	22

Figure 7: Concentration and Mass-activity of the most common primordial radionuclides in different types of rocks and soils (Ref.[8])

Nuclide	Historical Name	Half-life	Major Radiations
^{238}U	Uranium I	4.47×10^9 y	$\alpha, < 1\% \gamma$
^{234}Th	Uranium X ₁	24.1 d	$\beta,$
^{234}Pa	Uranium X ₂	1.17 m	$\beta, < 1\% \gamma$
^{234}U	Uranium II	2.46×10^5 y	$\gamma, < 1\% \gamma$
^{230}Th	Ionium	7.54×10^4 y	$\gamma, < 1\% \gamma$
^{226}Ra	Radium	1600 y	α, γ
^{222}Rn	Emanation	3.82 d	$\alpha, < 1\% \gamma$
^{218}Po	Radium A	3.10 m	$\alpha, < 1\% \gamma$
^{214}Pb	Radium B	26.8 m	β, γ
^{214}Bi	Radium C	19.9 m	β, γ
^{214}Po	Radium C	164.3 μ s	$\alpha, < 1\% \gamma$
^{210}Pb	Radium D	22.3 y	β, γ
^{210}Bi	Radium E	5.01 d	β
^{210}Po	Radium F	138.4 d	$\alpha, < 1\% \gamma$
^{206}Pb	Radium G	Stable	None

Figure 8: Uranium 238 decay chain characteristics, excluding minor branches (<<1% not shown) (Ref.[11])

Nuclide	Historical Name	Half-life	Major Radiations
^{232}Th	Thorium	$1.41 \times 10^{10} \text{ y}$	$\alpha, <1\% \gamma$
^{228}Ra	Mesothorium I	5.75 y	$\beta, <1\% \gamma$
^{228}Ac	Mesothorium II	6.15 h	β, γ
^{228}Th	Radiothorium	1.91 y	α, γ
^{224}Ra	Thorium X	3.66 d	α, γ
^{220}Rn	Emanation	55.6 s	$\alpha, <1\% \gamma$
^{216}Po	Thorium A	0.145 s	$\alpha, <1\% \gamma$
^{212}Pb	Thorium B	10.64 h	β, γ
^{212}Bi	Thorium C	1.01 h	α, γ
$^{212}\text{Po} (64\%)$ $^{208}\text{Tl} (36\%)$	Thorium C' / Thorium C''	0.300 ms / 3.05 m	$\alpha / \beta, \gamma$
^{208}Pb	Thorium D	Stable	None

Figure 9: Thorium 232 decay chain characteristics, excluding minor branches ($<1\%$ not shown) (Ref.[11])

Nuclide	Historical Name	Half-life	Major Radiations
^{235}U	Actinouranuim	$7.04 \times 10^8 \text{ y}$	α, γ
^{231}Th	Uranium Y	1.06 d	β, γ
^{231}Pa	Protoactinium	$3.28 \times 10^4 \text{ y}$	α, γ
^{227}Ac	Actinium	21.77 y	$\beta, <1\% \gamma$
$^{227}\text{Th} (98.62\%)$ $^{223}\text{Fr} (1.38\%)$	Radioactinium / Actinium K	18.72 d / 22.0 m	$\alpha, \gamma / \beta, \gamma$
^{223}Ra	Actinium X	11.44 d	α, γ
^{219}Rn	Actinon	3.96 s	α, γ
^{215}Po	Actinium A	1.78 ms	$\alpha, <1\% \gamma$
^{211}Pb	Actinium B	36.1 m	β, γ
^{211}Bi	Actinium C	2.14 m	α, γ
^{207}Tl	Actinium C'	4.77 m	$\beta, <1\% \gamma$
^{207}Pb	Actinium D	Stable	None

Figure 10: Uranium 235 decay chain characteristics, excluding minor branches ($<1\%$ not shown) (Ref[11])

1.2.2 Waste Origin

Now that all the main radionuclides involved into the NORM and TENORM waste radio toxicity were characterized, we examine the main ways these type of waste are produced.

The analysis will be made separating every sector from the different one.

- **Mining :**

- HARD ROCK METAL MINING : In the extraction and manufacturing process, some metals are strictly related to the natural radioactive isotopes. One example is Zircon (the zirconium silicate), which is found containing NORM material, prevalently uranium and thorium, which bonds form during the crystallization process, and then are very difficult to break. TENORM waste are produced in the moment when zircon is separated with other elements in an electromagnetic process, incrementing the radionuclides concentration. A similar discussion can be made for Titanium, which is the 9th most abundant element in the hearth crust, and that is found in the same sands in which can be found also radium, thorium and uranium, and due to this during the separation process, dust with an high level of radio toxicity can be formed;
- RARE EARTHS MINING WASTES : The extraction of rare earths mining wastes, which are around 15 chemical elements with atomic numbers between 57 and 71, known as lanthanides and actinides (making the exception for Yttrium39 and Scandium21), and which are used, thanks to their high electrical conductivity, in defense and modern electronics (for ex. To do magnets or chips), needs to be processed in minerals and ores containing uranium and thorium, in addition to potassium, which are removed, forming an increasing quantity of TENORM waste;
- BAUXITE and ALUMINA PRODUCTION : The most common way to produce Alumina, nowadays continue to be the Bayer Process, which is a process where Bauxite is dissolved in sodium hydroxide at high temperature and pressure, and then the formed alumina-bauxite ores are separated washing out the waste generated in the process, which are red muds, or sands. These red muds-sands have an higher percentage of uranium, thorium and radium progeny. (Although the radioactivity is not the primary concern, the main problem stays not in the specific radio toxicity of the waste, but in its quantities : for the production of 1ton of aluminum are produced nearby 2.5tons of solid waste which can contain different amounts of TENORM);
- COPPER EXTRACTION and PRODUCTION : The frequently occurring radionuclides which are contained into the minerals from

which copper is extracted, are mainly Uranium and Radium, and the mining and processing methods usually lead to concentrate and expose NORM, producing in this way TENORM waste. Due to the large scale of copper mining waste, a huge quantity of TENORM waste is generated by this sector. Radionuclides are concentrated and/or exposed during the copper processing, in different ways: during the leaching and solvent extraction may be extracted and concentrated radionuclides generating up to 2 level of magnitude over the background radiation; the minerals separated during the process (pyrite and sulfides) could be put in tailing piles, and be susceptible to leaching of radionuclides, in addition to this if exposed to water or air they could form acid mobilizing metals such as uranium (highly soluble in acid).

- URANIUM MINING : Uranium surface mining generates large volume of overburden with ambient or greater concentrations of uranium and its decay products. In the years due to the more efficient techniques that were gained, less accessible and lower grade ores were explored, increasing the ratio between overburden to ore level up to 60:1, from a starting 10:1 in the 50's. The most relevant radionuclide remains to be Radium226 in this case, for ex in a 1989 survey was measured a concentration in uranium mine overburden of Ra226 to be 0.9kBq/kg (Ref.[8]).

- **Energy Production :**

- OIL and GAS PRODUCTION : The production of TENORM waste in this sector is increasing both in radioactivity and volumes, due to the new extraction methods enhanced, specially the 'fracking' practice. In this sector can be easily found almost all the radioactive elements listed above, in addition to the U,Th, and Ra decay chain, there are also large quantities of K40 and of Pb and Po210. The waste are created at different level of the extraction process, in fact they can be: minerals inside the pipes, sediments, contaminated equipment and produced waters. In these waste the radioactivity level and the elements changes a lot from one to another; the sludge formed by dissolved solids precipitated from water contains higher levels of Radium 226 and 228 than others element, with an estimated concentration of them of 2.78 Bq/g, and can be even higher, representing in this way a high risk of exposure; the scale represent another problem from the radiation protection point of view in this sector, in fact approximately result to be produced 100 tons of scale per oil well every year, and in this waste there are high concentration of radionuclides : radium has been estimated to be 17.76Bq/g, but even much higher (were registered levels up to 14800Bq/g); In the particular extraction of gas, there is a prevalence of contamination from Rn222 to the entire equipment (pipes, valves,...), and other short lived radionuclides

like Bi214 can pose and be dangerous for the workers, and in these cases are of concern in long term point of view the quantities of Pb and Po 210 posed and generated in the equipment. Due to this high level of radiations almost all the waste generated are isolated : sludge is dewatered and held in storage tanks; waters are generally cleaned and reinjected into deep wells or reused; pipes are cleaned from the scale and if possible recycled, while the scale itself is placed in drums for later disposal; irradiated equipment if possible is recycled, if not is sent to landfills to be licensed as a TENORM waste.

- COAL COMBUSTION : Coal combustion generates residual containing concentrated NORM, so it's classified as a TENORM; the most frequent radionuclide found in these residuals are U,Th,Ra, and K. Radioactive materials are in fact comprehended into the composition of coal which is combusted during the energy production process, and at the end roughly the 10/20% of the total amount of ash and slag generated consists in TENORM waste. Obviously the conformation of coal changes from one type, to the other, and the coal combustion residuals (called CCR) can variate a lot from a deposit to another. The CCR can contain TENORM waste with concentration over 5 times the average background level. Here a picture of the different conformation of coal radionuclide composition depending on its origin.

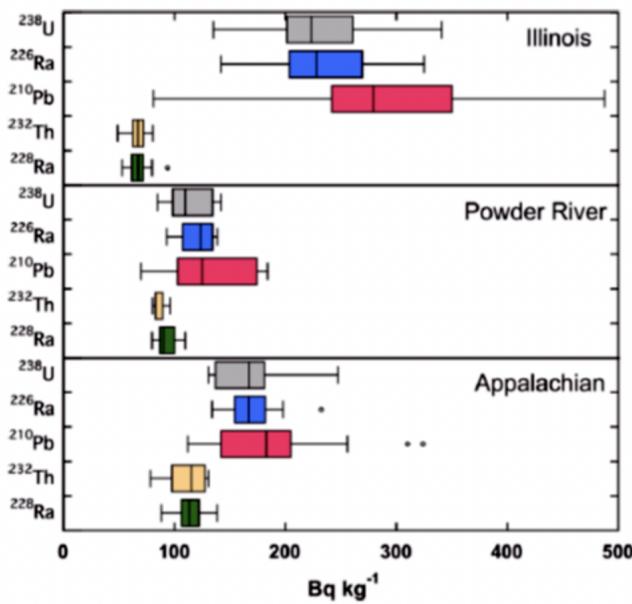


Figure 11: Coal different Radionuclides composition (Ref.[14])

In the fly ash and bottom ash, is estimate a value of Ra226 of about

0.14kBq/kg; Rn emanation could be a problem in the use of the ash as a material to produce concrete, but due to the lower emanation from the other glassy materials, the exposure at the end result to be mitigated. Typically the ash/residual which is not reused as a building material, is disposed in landfills or surface impoundments, for which there are regulations to follow.

- **Water Treatment :**

- DRINKING WATER RESIDUALS : due to the fact that primordial radionuclides are present in every rock/soil, they can accumulate in drinking water sources. As the water is treated, and the soluble radionuclides (U, Ra and Rn above all) are present, then is very likely to have a higher concentration of radionuclides collected in sediments, sludge, filters, tanks, and so on, creating TENORM waste. There are different types of treatment used to clean water, which accumulate in the process the different radionuclides : ‘alum treatment’, where alum is introduced as a gel to bond with different particles (in which we have radionuclides also), and gradually is collected in the bottom of the treatment tanks; ‘lime softening’ is a process used to soften water with the addition of calcium hydroxide, that has the effect of raising water pH, and forming a solid sludge in which the 90% of Ra is trapped, and then removed, and usually these sludges reach levels of 0.2-1.1kBq/kg; ‘Ion exchange’ is another particular process, which consists in replacing calcium (Ca^{2+}) and magnesium (Mg^{2+}) ions with the sodium ones (Na^+), removing in this way 95% of Ra, and also removing U, in this way the form ion-exchange resins with Ra^{226} concentrations up to 3700kBq/kg ; ‘reverse osmosis’ is also used, and like the name suggests are used membranes to clean up water, and doing this higher concentrations of radionuclides can build up. The disposal of the TENORM waste in this sector variates a lot from one process to the other : the alum sludge generated is usually disposed in storage ponds, with the concentration of Ra and U periodically controlled; contaminated materials aree also disposed in landfills, usually compacted and covered, ensuring the prevention of Rn to leach in the groundwater.
- WASTEWATER TREATMENT : water discharge from civilian use can form TENORM waste as large volumes are collected to be treated, and these volume comprehend large volumes of radionuclides naturally occurring or man-made, which depending on sludge/ash produced composition need to be treated and then disposed in different ways. Under we have two tables from Ref.[15]: on the left the the average concentration of radionuclides in sludge, and on the right the average concentration of radionuclides in ash.

Treatment Technology	Contaminant Removed	Removal Efficiency	Wastes Produced	Waste Concentrations
Cation exchange	Radium	85-97%	Rinse & backwash water Regenerant brine	8 to 94 pCi/L-Ra ¹ 50 to 3,500 pCi/L-Ra ¹ 22 to 94 pCi/L ²
Anion exchange	Uranium	95%	Rinse & backwash water Brine regenerant solution	2 to 6e+06 pCi/L-U 35 to 4.5e+06 pCi/L-U 1.3 to 11 pCi/L
Lime softening	Radium Uranium	90% 85-90% ³	Sludge (at clarifier) Sludge (dry) Filter backwash	76 to 4,577 pCi/L-Ra 1 to 21.6 pCi/g-Ra 1 to 10 pCi/g-U 6.3 to 21.9 pCi/L-Ra
Reverse osmosis	Radium Uranium	90+% ---	Reject water	7 to 43 pCi/L-Ra 200 to 750 pCi/L-U
Electrodialysis	Radium Uranium	90% ---	Reject water	No data
Iron removal -Oxidation -Greensand	Radium	0 to 70% ⁴	Solids & supernatant from filtration backwash Green sand Media	12 to 1,980 pCi/L-Ra 28 to 250 pCi/g-Ra
Selective sorbents	Radium Uranium	90+%	Selective sorbents (radium selective and activated alumina)	up to 3.6 pCi/g-Ra
Coagulation/ Filtration	Uranium	50 to 85%	Sludge	10,000 to 30,000 pCi/L-U

Figure 12: Technology and associated waste produced for the treatment of water (Ref[15])

Radionuclide	All concentrations are expressed in pCi/g dry, unless noted: 1 pCi/g=37 Bq/kg.				Radionuclide	All concentrations are expressed in pCi/g dry, unless noted: 1 pCi/g=37 Bq/kg.				
	Min	Median	95th P	No. Detects		Minimum	Median	95th P	Maximum	
Alpha	ND	7	34	137	309/311	5	27	93	178	
Beta	1.7	1.9	24	37	309/311	15	15	144	253	
Am-241	ND	1.2	ND	2.5	10/311	ND	ND	0.21	2/35	
Bk-7	ND	ND	9	22	26/311	ND	4.9	30	24/35	
Bk-242	ND	ND	3.3	3	15/311	ND	1.2	3.5	15.7	
Bi-214	0.3	2.3	16	16	2/30/311	ND	2.4	14	34/35	
Bi-214	ND	ND	1	0.08	1/311	ND	ND	ND	1	
Ca-41	ND	ND	ND	0.016	1/311	ND	ND	ND	0/35	
Co-57	ND	ND	ND	0.26	6/311	ND	ND	ND	0/35	
Co-60	ND	ND	1	0.11	13/311	ND	ND	ND	2/35	
Cr-51	ND	ND	ND	3.5	6/311	ND	ND	ND	0/35	
Cs-134	ND	ND	ND	0.04	1/311	ND	ND	ND	0/35	
Cs-137	ND	ND	0.11	9.6	133/311	ND	ND	ND	0/35	
Eu-154	ND	ND	ND	0.01	1/311	ND	ND	ND	0/35	
Fm-257	ND	ND	ND	0.4	1/311	ND	ND	ND	0/35	
H-3	0.3	5	8	111/158	ND	ND	ND	ND	0/35	
I-129	ND	ND	ND	0.01	1/311	ND	ND	ND	0/35	
I-131	ND	1.8	51	840	246/311	ND	ND	ND	0/35	
In-111	ND	ND	0.04	3.6	18/311	ND	ND	ND	0/35	
In-113	ND	ND	ND	0.01	1/311	ND	ND	ND	0/35	
La-138	ND	ND	ND	0.07	1/311	ND	ND	ND	0/35	
La-138	ND	ND	ND	0.26	30/311	ND	ND	ND	0/35	
Pb-210	ND	ND	4	27	96/311	ND	ND	ND	0/35	
Pb-212	ND	ND	4.4	1.9	15	30/311	ND	ND	0/35	
Po-210	ND	ND	ND	0.11	20/311	ND	ND	ND	0/35	
Po-214	ND	ND	ND	0.01	1/311	ND	ND	ND	0/35	
Po-238	ND	0.01	0.07	0.19	75/92	ND	0.015	0.1	0.1	
Po-238	ND	ND	ND	0.04	6/92	ND	ND	ND	2/28	
Rs-223	ND	ND	0.09	2/311	ND	ND	ND	ND	0/35	
Rs-224	ND	ND	0.9	12	47/311	ND	ND	ND	0/35	
Rs-225	ND	ND	5	53	298/311	ND	ND	ND	0/35	
Rs-225	ND	0.82	6.1	38	275/311	ND	ND	ND	0/35	
Rs-226	ND	ND	ND	0.01	1/311	ND	ND	ND	0/35	
Sm-153	ND	ND	ND	27	130/311	ND	ND	ND	0/35	
Si-39	ND	ND	0.35	20	70	64/98	ND	ND	0/35	
Si-39	ND	ND	ND	1.1	1	4/45	ND	ND	0/35	
Th-227	ND	ND	0.1	0.5	49/207	ND	ND	ND	0/35	
Th-230	ND	ND	0.01	0.001	4/311	ND	ND	ND	0/35	
Th-230	0.09	0.34	1	1.7	92/92	ND	0.75	2.3	2.6	
Th-232	0.02	0.2	0.6	1.6	92/92	ND	0.605	1	1.7	
Th-232	ND	ND	0.8	1.7	151/311	ND	ND	ND	0/35	
Tl-201	ND	ND	48	241	151/311	ND	0.62	79	105	
Tl-201	ND	ND	ND	0.53	1/311	ND	ND	ND	0/35	
Tl-208	ND	ND	0.07	0.96	4.8	160/311	ND	0.66	2.3	13.8
U-234	0.18	1.96	17	44	92/92	1.2	5.55	49	91	
U-235	ND	ND	0.48	1	151/311	ND	0.15	3.4	23.5	
U-238	0.18	1.4	12	26	92/92	0.8	3.3	35	74	
Zn-65	ND	ND	ND	0.06	1/311	ND	ND	ND	0/35	

Indicates concentrations for this radionuclide are expressed in pCi/g wet.

Figure 13: Average concentration of radionuclides in sludge (left), and in ash (right) (Ref.[15])

● Consumer Products :

Some TENORM can be found also in certain consumer products (even if with time, due to the increasing attention from the radiological point of view, these material are less and less). Is not unlikely to have radioactive antiques at home without knowing their spreading of radiations (like said above they could have been sold during the last century, when the effect of radiations, and their study was not already deepened).

- TOBACCO PRODUCTS : represent the largest radiation doses received in the public after the Rn. Tobacco plants tend in fact to absorb the radioactive elements from the soil, and also to accumulate Rn decay products in the tobacco leaves (above all Po210), which then are inhaled by consumers while smoking;

- FERTILIZER : the fertilizers are mainly formed by phosphate and phosphate rocks, materials that could contain notable amounts of radioactive material, which is then processed, increasing in this way the concentration of radionuclides, and creating TENORM materials. Phosphate fertilizers are not considered radioactive materials, even though they could contain up to 1.11Bq/g of Ra226; the real concern is given to the production of waste created during both the thermal and wet processes in the fertilizer production : the wet process is characterized by the production of phosphogypsum (a solid-liquid mixture of phosphoric acid and calcium sulfate) as a waste, this time the problem is not given by the radio toxicity itself of the material (also Ra226 up to 1.3Bq/g and U up to 3.7Bq/g), but by the fact that this type of waste is produced in very large quantities, and with time is becoming more and more difficult to manage these large piles in land or river (also during time there could be more problems related to the easy production and leaching of Rn from the ‘gyp stacks’); then there’s the thermal process, whose principal produced waste results to be the phosphate slag, a sort of glassy material formed in the furnace, which could contain up to 1.85 Bq/g of both Th and Ra, but is even less worrying than the phosphogypsum being characterized by a littler leaching probability of Rn. Is important also to underline that low volume of discrete TENORM waste can be produced in wet-plants, into the pipes : in fact the scale can reach in these case up to 3700kBq/kg;
- BUILDING MATERIALS : being formed by rocks also the building materials are subject to different levels of concentration of natural radionuclides. Surely for humans the most dangerous elements in the decay pathways remain to be Rn, that if inhaled in big quantities can lead to cancer. Even if the released levels of Radon are not as relevant as the ones that could come from the soil, the presence at home of materials like quartz and granite would cause an increase of annual radiation dose above normal background level.

1.2.3 Conclusions

TENORM waste represent unique problems from the radiological point of view because of their large volumes of production in which they are cumulated. They are all characterized by long-live radionuclides (all listed at the beginning), and due to this, they have to be monitored in order to not exceed volumes above which the radio toxicity could become a serious problem. Mainly the problems are given by the Radium concentration in the materials, which can lead to the formation to Rn222, which represent certainly the most dangerous natural radionuclide for human kind in the ones of primordial formation. From the disposal point of view almost all the waste in the mining sector listed here, and also in the consumer products and water treatment don't need particular treatment and could be disposed in landfills either buried or not, depending on the activity level and on the regulations; different speech if we talk about the oil and gas production, coal combustion and also some particular waste (but in lower volume) of the other categories listed above, where we can have relatively high levels of radionuclides, with long half-lives, and due to this is necessary to dispose them in protect environments to avoid possible public exposures to ionizing radiations, being catalogued either in LLW, or ILW, depending on the safety authorities and on the disposal site. Definitely these type of waste surely don't represent an important part of HLW which needs long term isolated disposal due to their, in general, low specific activity.

1.3 Medical Waste Characterization

Making an analysis of the Radioactive waste sector, with a particular eye on the HLW production and their isotopic composition, it's very important to give a background of the use of radionuclides in medicine, due to the importance that this branch has reached nowadays. In this part will be briefly shown the main uses of both sealed and unsealed radioactive sources, the main differences in applications and above all what are the most used radionuclides used in medicine. Will be made a distinction with the three branches of medicine in which they are used: diagnostic, therapy and research.

1.3.1 Radionuclides Characterization (and Uses)

Radioactive materials can be of two different categories in Nuclear Medicine : **Unsealed Radionuclides , Sealed Radionuclides.**

Here follows a description of these two different categories.

- **Unsealed Radionuclides :**

- Diagnosis

The use of radionuclides for diagnostic purposes has both “in vitro” and “in vivo” applications. In vitro means studies performed on human biological samples outside of the human body while in vivo refers to dynamic function studies within the human body. In vitro applications typically involve kBq activities of aqueous-based radionuclides being utilized to measure levels of drugs or hormones in biomedical samples. Typical radionuclides include ^{125}I , ^{57}Co ^{58}Co and ^{14}C . By far the greatest diagnostic application of radionuclides in medicine is in vivo investigation of body function using gamma camera imaging. Many in vivo radio-pharmaceuticals are prepared by diluting a pharmaceutical with $^{99\text{m}}\text{Tc}$, which is eluted from a $^{99\text{m}}\text{Tc}$ generator. Radionuclides are also administered in vivo to act as tracers in monitoring body functions. The usual range of administered doses for technetium radio-pharmaceuticals is 40–800 MBq, with lower doses administered for pediatric patients. Other common diagnostic imaging radionuclides include: ^{67}Ga , ^{111}In , ^{201}Tl , ^{123}I and ^{131}I . These radionuclides are usually administered at activity levels in the range of 40–400 MBq for imaging purposes. Some radionuclides are also used to label human blood components to act as tracers for sites of blood loss or sites of infection. This typically involves removing a blood sample from the patient, radiolabelling the blood and re-injection. Examples of the radionuclides used include $^{99\text{m}}\text{Tc}$, ^{111}In , ^{51}Cr , ^{59}Fe and ^{125}I . The actual activity that may be re-injected is usually in the range of a few MBq to a maximum of 200 MBq, with the highest activity typically used for $^{99\text{m}}\text{Tc}$. Radioactive gases and aerosols are used for diagnostic purposes during lung ventilation

imaging. This involves the use of ^{81}mKr (up to 6 GBq administration per patient), ^{133}Xe (up to 400 MBq) and ^{99}mTc -diethyl tetra penta acetic acid (DTPA) aerosol inhalation (up to 80 MBq inhalation activity);

- Therapy

Therapeutic applications of radionuclides in medicine utilize a number of unsealed sources in much higher activities than those used for diagnosis. Iodine-131 is widely used for treatment of thyrotoxicosis and for ablation of the thyroid tissue or metastases during cancer treatment. Individual patient doses are typically in the range of 200 MBq — 11 GBq. The ^{131}I used for therapeutic purposes may be provided in three physical forms — liquid sodium iodide for dispensing as multiple individual patient doses for oral administration, individual powder filled gelatin capsules for oral administration or sterile sodium iodide solution for injection. Injections are normally only administered where there may be a problem with oral administration. Use of other therapeutic unsealed radionuclides usually involves venous injection of a sterile, undiluted solution of the radionuclide, e.g. ^{89}Sr or ^{32}P . Strontium is typically used in therapy for the management of pain associated with bone metastases. Administered doses are usually several hundred MBq. Yttrium-90 is typically injected into the joints of a patient, e.g. knee, as a silicate colloidal solution, with administered activity levels of about 200 MBq per injection. Some radionuclides used in therapeutic applications are diluted prior to administration. This practice may increase the volume of wastes requiring further management. For example, therapeutic administration of ^{131}I -MIBG (meta-iodo benzyl-guanidine) is usually diluted with sterile isotonic saline and is intravenously administered slowly over a period of up to an hour or more using a pump system. This results in the generation of additional solid radioactive waste, such as disposable plastics;

- Research

A wide range of unsealed radionuclides are used for research purposes both in health care biological research centers and pharmaceutical development facilities [1]. Isotopes such as ^{3}H , ^{35}S , ^{32}P and ^{33}P are widely used for DNA sequencing in research. The range of radionuclides used in biomedical research is much wider than the number of radionuclides used for in vitro and in vivo diagnostic/therapeutic purposes. The reason for this is that a large number of new uses of radionuclides are often evaluated for several years in animal studies at pharmaceutical development establishments prior to being approved for use. This is necessary to verify that the product is safe for human administration. Wastes generated as part of drug evaluation research may often be stored frozen for prolonged periods (2–5 years) on the user's premises whilst the drug regulatory organizations evaluate the

results of clinical trials. The final approval of the drug for use can involve the sudden requirement for disposal of large volumes of frozen wastes, including animal carcasses, tissues, organs, blood products, urine and faeces and by-products of radiolabelling experiments. The use of radionuclides such as ^3H and ^{14}C in GBq quantities for organic synthesis is not uncommon in pharmaceutical research, resulting in low volume, high activity waste for management and disposal. Often the waste generated from biomedical research is more difficult to control due to transient workers on research grants or project work by students. Research work typically involves the use of animals, hence wastes will include animal tissues and carcasses, contaminated bedding material and excrement. Often the wastes from these sources are more difficult to manage and to quantify;

Radionuclide	Half-life	Principle application	Typical quantity per application	Waste characteristics
^3H	12.3 a	Radiolabelling Clinical measurement Biological research Organic synthesis	Up to 50 GBq	Solvents, solid liquid
^{13}N	10 m	Positron emission tomography	Up to 2 GBq	Solid, liquid
^{11}C	20.4 m	Positron emission tomography	Up to 2 GBq	Solid, liquid
^{14}C	5730 a	Medical diagnosis Biological research Labeling	Less than 1 MBq Up to 50 GBq Up to 50 GBq	(Exhaled CO ₂) Solid, liquid Solvent
^{15}O	122 s	Positron emission tomography	Up to 500 MBq	Solid, liquid
^{18}F	1.8 h	Position emission tomography	Up to 500 MBq	Solid, liquid
^{22}Na	2.605a	Medical diagnosis	Up to 1MBq	Solid, liquid
^{24}Na	15.0 h	Biological research	Up to 5 GBq	Liquid effluent
^{32}P	14.3 d	Clinical therapy	Up to 200 MBq	Solid, liquid
^{33}P	25.4 d	Biological research	Up to 50 MBq	Effluent
^{35}S	87.4 d	Medical and biological research	Up to 5 GBq	Solid, liquid effluent
^{36}Cl	3.01×10^5 a	Biological research	Up to 5 MBq	Gaseous, solid, liquid
^{39}K	7.6 m	Positron emission tomography	Up to 1 GBq	Solid, liquid
^{42}K	12.4 h	Clinical measurement	Up to 5 MBq	Solid, liquid
^{43}K	22.2 h	Clinical measurement	Up to 5 MBq	Solid, liquid
^{45}Ca	163 d	Biological research	Up to 100 MBq	Mainly solid,
^{45}Ca	4.54 d	Medical diagnosis	Up to 100 MBq	some liquid
^{46}Sc	83.8 d	Medical and biological research	Up to 500 MBq	Solid, liquid
^{51}Cr	27.7 d	Clinical measurements Biological research	Up to 5 MBq Up to 100 MBq	Solid Mainly liquid effluent
^{57}Co	271.7 d	Clinical measurements	Up to 50 MBq	Solid, liquid
^{58}Co	70.8 d	Biological research	Up to 5 MBq	Effluent
^{59}Fe	44.5 d	Clinical measurements Biological research	Up to 50 MBq	Solid, mainly liquid effluent
^{67}Ga	3.3 d	Clinical measurements	Up to 200 GBq	Solid, liquid effluent
^{68}Ga	68.2 m	Positron emission tomography	Up to 2 GBq	Solid, liquid
^{67}Cu	2.6 d	Clinical therapy Monoclonal antibodies	Up to 1 GBq	Solid, liquid
^{75}Se	119.78 d	Clinical measurements	Up to 10 MBq	Solid, liquid
^{75}Br	98 m	Medical diagnosis		Solid, liquid
^{76}Br	16.2 h	Medical diagnosis		Solid, liquid
^{75}Br	57 h	Clinical measurement	Up to 5 MBq	Solid, liquid
$^{81\text{m}}\text{Kr}$	13.3 s	Lung ventilation studies	Up to 6 GBq	Gaseous
^{82}Rb	76 s	Positron emission tomography		Solid, liquid
$^{82\text{m}}\text{Rb}$	6.2 h	Clinical measurement		Solid, liquid
^{86}Rb	18.7 d	Medical and biological research	Up to 50 MBq	Solid, liquid

Figure 14: Radionuclides used in Medicine in Unsealed form (Ref.[16])

⁸² Sr	64.8 d	Medical diagnosis/research	Up to 50 MBq	Solid, liquid
⁸⁵ Sr	50.5 d	Clinical therapy	Up to 300 MBq	Solid, liquid
⁹⁰ Y	2.7 d	Clinical therapy Medical and biological research	Up to 300 MBq	Solid, liquid
⁹⁵ Nb	35.0 d	Medical and biological research	Up to 50 MBq	Solid, liquid
^{99m} Tc	6.0 h	Clinical measurements Biological research Nuclide generators	Up to 100 GBq	Solid, liquid
¹¹¹ In	2.8 d	Clinical measurements Biological research	Up to 50 MBq	Solid, liquid
¹²³ I ¹²⁴ I ¹²⁵ I ¹³¹ I	13.2 h 4.2 d 60.1 d 8.0 d	Medical and biological research Medical diagnosis/research Clinical measurements Clinical therapy	Up to 500 MBq Up to 11.1 GBq	Solid, liquid Occasionally vapor
¹¹³ Sn	155.0 d	Medical and biological research	Up to 50 GBq	Solid, liquid
¹²⁷ Xe	36.4 d	Medical diagnosis	Up to 200 MBq	Gaseous, solid
¹³³ Xe	5.3 d	Clinical measurements	Up to 400 MBq	Gaseous, solid
¹⁴¹ Ce	32.5 d	Medical research	Up to 50 MBq	Solid, liquid
¹⁵³ Sm	47 h	Clinical therapy	Up to 8 GBq	Solid, liquid
¹⁶⁹ Er	9.3 d	Clinical therapy, palliative treatment	Up to 500 MBq	Solid, liquid
¹⁸⁶ Re	3.8 d	Clinical therapy, palliative treatment	Up to 500 MBq	Solid liquid
¹⁸⁸ Re	17 h	Clinical therapy	Up to 500 MBq	Solid, liquid
¹⁹⁸ Au	2.7 d	Clinical measurements, therapy	Up to 500 MBq	Solid, liquid
²⁰¹ Tl	3.0 d	Clinical measurements	Up to 200 MBq	Solid, liquid
²⁰³ Hg	46.6 d	Biological research	Up to 5 MBq	Solid, liquid

Figure 15: Radionuclides used in Medicine in Unsealed form (cont.) (Ref.[16])

• Sealed Radionuclides :

– Diagnosis

Sealed radiation sources may be used for the following diagnostic purposes:

- * 3/4 bone densitometry;
- * 3/4 anatomical marking;
- * 3/4 calibration and reference standards.

Density scanners for bone mineral determination are one example of a diagnostic application of radiation sources in medicine. Typical sources used for these purposes are ²⁴¹Am, ¹⁵³Gd or ¹²⁵I at activity levels of up to several GBq. At the end of their useful life, these spent radiation sources should be sent to a centralized facility for treatment and disposal. Since anatomical markers, calibration sources and reference standards may have small dimensions and low activity, special care should be taken to ensure that they are not lost in use, e.g. accidentally discarded with the normal waste or misplaced during medical applications;

– Therapy

A number of different radionuclides are used in the form of sealed sources for clinical treatment during manual brachytherapy, remote after-loading brachytherapy, tele-therapy, blood irradiation and other purposes. Since sources of rather high activity may be involved, attention should be paid to proper shielding, storage and security as soon as the sources are taken into use. Sealed sources are used in a wide range of activities for therapeutic purposes. Many are directly implanted during oncology treatments or applied to a patient, e.g.

¹⁰⁶Ru eye plaques and implants of ¹⁹²Ir, ¹³⁷Cs and ¹⁹⁸Au. Sealed sources are also now being used in vascular treatments for stenosis as a complement to angioplasty during catheterization. Sources include both beta and gamma emitters such as ¹⁹²Ir and ⁸⁹Sr. Larger sealed sources such as ⁶⁰Co are used in tele-therapy heads for beam treatments of malignant conditions. Cobalt-60 is also used in gamma knife surgery where approximately 200 sources are focused on a very small portion of the patients head. These applications may involve activities of up to several hundred TBq. Some therapeutic sealed sources are not used directly for human therapy, e.g. the TBq sources of ¹³⁷Cs or ⁶⁰Co used in blood cell irradiators. Although radium sources are no longer used in good medical practice, spent sources may still be stored and require treatment and disposal. Specific precautionary measures should be applied to their storage because such sources will eventually leak;

- Research

Sealed radioactive sources may also be used in research and teaching/training establishments. These may involve small sealed calibration sources used for gamma and liquid scintillation counting and ⁶³Ni sources for gas chromatography. Some establishments may also use much higher activity sealed sources in irradiators such as ¹³⁷Cs or ⁶⁰Co sources.

Application	Radioisotope	Half-life	Source activity	Comments
Bone densitometry	²⁴¹ Am ¹⁵³ Gd ¹²³ I	433.0 a 244.0 d 60.1 d	1–10 GBq 1–40 GBq 1–10 GBq	Mobile units
Manual brachytherapy	¹⁹⁸ Au ¹³⁷ Cs ²²⁶ Ra ⁶⁰ Co ⁹⁰ Sr ¹⁰³ Pd ¹²⁵ I ¹⁹² Ir ¹⁰⁶ Ru ⁹⁰ Y	2.7 d 30.0 a 1600 a 5.3 a 29.1 a 17.0 a 60.1 d 74.0 d 1.01 a 2.7 d	50–500 MBq 30–300 MBq 50–500 MBq 50–1500 MBq 50–1500 MBq 50–1500 MBq 200–1500 MBq 5–100 MBq 10–20 MBq 50–500 MBq	Small portable sources
Vascular brachytherapy	³² P ⁸⁹ Sr ¹⁹² Ir	14.3 d 50.5 d 74 d	200 MBq 150 MBq 0.1–1 TBq	Catheterization
Remote after loading brachytherapy	¹³⁷ Cs ¹⁹² Ir	30.0 a 74.0 d	0.03–10 MBq 0.1–200 TBq	Mobile units
Teletherapy	⁶⁰ Co ¹³⁷ Cs	5.3 a 30.0 a	50–1000 TBq 500 TBq	Fixed installations
Whole blood irradiation	¹³⁷ Cs ⁶⁰ Co	30.0 a 5.3 a	2–100 TBq 50–1000 TBq	Fixed installations
Research	⁶⁰ Co ¹³⁷ Cs	5.3 a 30.0 a	Up to 750 TBq Up to 13 TBq	Fixed installations
Calibration sources Anatomical markers Sources as standards in instruments	⁶³ Ni ¹³⁷ Cs ⁵⁷ Co ²²⁶ Ra* ¹⁴⁷ Pm ³⁶ Cl ¹²⁵ I	96 a 30.0 a 271.7 d 1.6×10^5 a 2.62 a 3.01×10^5 a 1.57×10^7 a	<4 MBq <4 MBq Up to 400 MBq <10 MBq <4 MBq <4 MBq <4 MBq	Fixed installations in instruments or mobile sources
Gamma radiosurgery knife	⁶⁰ Co	5.3 a	Up to 220 TBq	Skull cap

* Radium sources are no longer used for therapeutic treatment but exist as spent sources in some hospitals.

Figure 16: Sealed Sources used in Medicine (Ref.[16])

1.3.2 Types of Biomedical Radioactive Waste

The use of this wide range of radionuclides in medicine and medical research leads to the generation of waste, which requires a comprehensive management system. In many instances, the potential additional hazards, either from the chemical, biological or physical properties are greater than the radiological hazard due to the presence of radionuclide contamination. The following is a non-exhaustive list of the types of radioactive waste that may occur as a result of the use of radionuclides in medicine:

- surplus solutions of radionuclides from diagnostic, therapeutic and research applications which are likely to be sterile;
- aqueous based solutions containing low levels of radionuclides, e.g. from washing of apparatus;
- organic based solutions which may or may not be miscible with water, e.g. liquid scintillation counting residues and residues from organic synthesis;
- excreta from patients administered with radionuclides for diagnostic or therapeutic purposes;
- anatomical wastes, e.g. body parts, tissues, organs and fluids;
- spent radionuclide generators or spent radioactive solutions, such as those from radiopharmaceutical preparation;
- miscellaneous solid and semi-solid, wet wastes which may or may not be suitable for landfill or combustion, e.g. incontinence pads soiled with excreta, absorbed liquids;
- resin columns, matrix gels and chromatography plates from medical diagnosis and research;
- food waste from patients administered with radionuclides for therapeutic purposes, e.g. ¹³¹I ablation therapy;
- miscellaneous solid, dry wastes which are suitable for compaction, combustion or shredding, e.g. gloves, paper tissues;
- miscellaneous solid, dry wastes which may be not suitable for compaction, combustion or shredding, e.g. furniture and equipment parts;
- miscellaneous wastes which pose a puncture hazard, e.g. needles, broken glass, vials;
- waste from spills, decontamination and decommissioning procedures, e.g. liquids absorbed on matrix, mops, tissues;
- filters used in equipment, e.g. charcoal traps, fume hood filters;

- fragments from sources used in brachytherapy, e.g. cut lengths of iridium wire;
- ancillary wastes, e.g. materials which may have come into contact with humans or animals;

In this general overview we can also split the wastes in some categories that have to be managed in different ways:

- Liquid waste: liquid radioactive waste includes contaminated water and effluent, waste arising from chemical processing and decontamination solutions, solvents, blood or body fluids, discarded liquid radiopharmaceuticals, wound or oral discharges, urine, chemotherapy agents, small quantities of contaminated oils and scintillation fluids. Waste that includes both radioactivity and a hazardous chemical component is usually referred to as a mixed waste;
- Gaseous waste: Xenon-133 and 81mKr are used in diagnostic imaging for assessment of regional lung ventilation. Since they are noble gases, they are difficult to treat and are often released to the atmosphere through an exhaust system. It is essential to ensure that there is no possibility of re-entry of the released gases back into the building through open windows or ventilation system;
- Solid waste: at health care, medical and research facilities, solid waste is generated in the form of paper and plastic, animal carcasses, contaminated materials, discarded radiopharmaceutical containers, bandages, contaminated equipment or organs and tissues. Solid waste is typically classified as combustible/non-combustible and compactible/non-compactible waste. It generally contains a relatively low level of radioactivity when compared to liquid wastes. Solid radioactive waste consists mainly of general biomedical waste, which includes protective clothing, plastic sheets and bags, gloves, masks, filters, overshoes, paper wipes, towels, metal and glass, hand tools and discarded equipment. Spent sealed sources: sealed sources at the end of their useful clinical life are categorized as waste which needs to be properly conditioned and disposed of. Spent sources could be divided into the following categories:
 - * Sources with half-life ≥ 100 days, with high activity content such as ^{192}Ir (200– 1500 MBq);
 - * Sources of low activity used for calibration and as standards;
 - * Sources with a potential emanation and contamination hazard. Special security and radiological precautions need to be taken for the handling and the storage of spent radium sources and sources known to be leaking;
 - * Sources with half-life < 100 days, with low or high activity. Decommissioning waste: use of accelerators in medicine may also produce

radioactive waste, specifically during decommissioning of these facilities. The use of accelerators can create an activation problem of surrounding materials, particularly with neutrons of energy higher than 10 MeV.

1.3.3 Conclusions

Radioactive waste from the medical sector do not present a significant long term waste management problem when compared to other type of waste (for ex. the ones from nuclear fuel cycle operations, NPP, and so on...). The main characteristics of these biomedical waste are their short half-life and low radio-toxicity. Typically they have low energy β and γ decay and are generally of low total and specific activity. Important considerations on the other hand are the volumes of waste and other hazardous properties associated with the waste such as biological and chemical risks, which in almost all the cases could be considered the most dangerous one.

(Here a short summary of these types of non-radiological hazard :

- Physical hazards, which include the possibility of cuts and puncture injuries such as those from needles, broken glass, scalpel blades or blood lancets (sharps). They also include injuries sustained as a result of manual handling of heavy objects such as shielding, containers, radioactive patients, etc.;
- Chemical hazards, which include the potential for adverse chemical reactions or injuries which may be posed by the presence of acids, alkalis, oxidizers or oxidizable organic matter;
- Biological/infectious hazards, as any waste generated in a health care facility which is contaminated with human blood, other body fluids, or any potentially infectious material is determined as “biohazardous”;
- Flammable/explosive hazard, such hazards can arise when low flash point organic scintillants ($>21^{\circ}\text{C}$) are stored. (Nb. It is essential that the radioactive store has spark resistant lighting and that these wastes are securely stored in metal bins/cabinets. The radioactive store should also be adequately ventilated to prevent the buildup of fumes and a subsequent explosion hazard. The radioactive store should not be located in the vicinity of other flammable hazards such as compressed gases or highly combustible wastes.).)

As other types of long term nuclear waste, also the limited number of the ones coming from medical sector need a pre-treatment, a treatment and a disposal or reprocessing facility to be managed, but with waste that don't need such a process, because they are considered short term ones (composed by short-living radionuclides), only radioactivity level controls are practiced (after a period of storage): if these are below clearance level, they are managed as simple municipale wastes, after a process that involves disinfection, incineration and a

continuous control in quality. Hence, we can say that in general, except in some cases, radioactive wastes from medical field don't need a particular and complex technology of storage, and their management is easier than other sectors.

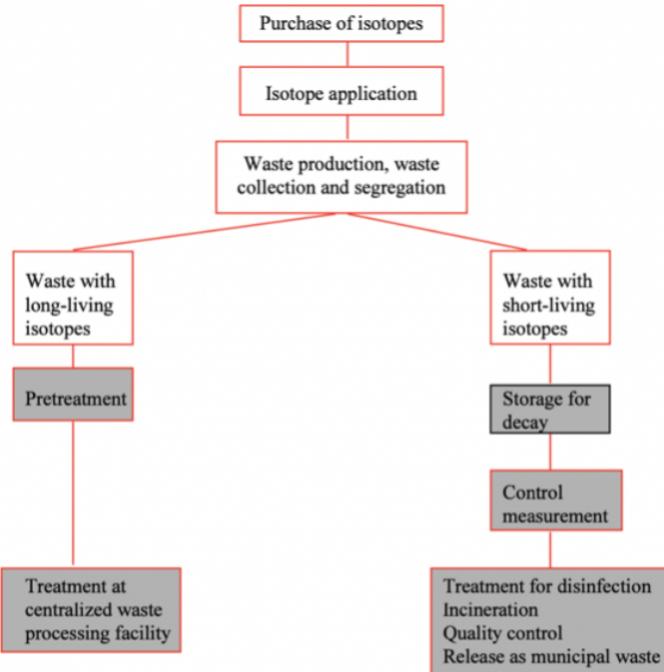


Figure 17: Scheme of the Management of Biomedical Radioactive Waste (Ref.[16])

1.4 Nuclear Reactors Related Waste Characterization

The waste material coming from Nuclear Reactors with both the Power Production, or the Research aim, form the most relevant part of radioactive material that every year needs to be disposed due to its high activity level and long live radionuclides (either from decommissioning or from operating plants). Due to this, in the following section there will be a deep analysis over this type of HLW, which, how will be explained in the end, have a central role in the materials that will be used for our Numerical Simulations.

1.4.1 Radionuclides Characterization

The aim of this paragraph is to characterize the radionuclides from Nuclear Reactors which are part of the composition of radioactive waste materials, giving more attention to the ones which are particularly relevant either from the radiological point of view, either from the size of waste generated, and that have to be managed while operating (ex. spent fuel discharge) or during the decommissioning period. It's very important while reading this part to take in mind that all the radionuclides are found in different concentration depending on the type of reactor considered, the time that had passed before the decommissioning, the site where they are placed, the management of the NPP, and so on...

In a more general vision, is possible to state that the most relevant radionuclides that can be found during the decommissioning of a nuclear power plant are listed here in the following table :

Isotopo	Tempo di dimezzamento (anni)	Modalità di decadimento principale	Energia dei γ eventualmente emessi (MeV)	Materiali ove sono possono essere presenti gli isotopi
${}^{3}H$	12,3	β^-	-	A, Al
${}^{14}C$	5730	β^-	-	G, Ac, Al
${}^{23}Na$	2,6	CE, β^+	0,51 - 1,28	A
${}^{30}Ar$	$3,1 \times 10^5$	β^-, CE	-	C
${}^{37}Ar$	269	β^-	-	C
${}^{40}Ca$	1×10^5	CE	-	C
${}^{45}Ca$	0,4	β^-	-	C
${}^{46}V$	0,9	CE	-	Al
${}^{54}Mn$	0,9	CE, γ	0,83	All, Ac, Al
${}^{57}Fe$	2,7	CE	-	C, Ac, A, Al
${}^{60}Co$	0,7	CE, γ	0,12 - 0,14	Al
${}^{60m}Co$	5,3	β^-, γ	1,2 - 1,3	C, Ac, A, Al, Z
${}^{58}Ni$	$7,5 \times 10^5$	CE	-	C, Ac, A, Al, Z
${}^{61}Ni$	100	β^-	-	C, Ac, A, Al
${}^{62}Zn$	0,7	β^-, γ, CE	0,51 - 1,12	Al
${}^{65}Cr$	39	β^-	-	Z
${}^{87}Rb$	$1,5 \times 10^5$	β^-	-	A, Z
${}^{95}Nb$	2×10^4	β^-, γ	0,70 - 0,87	Ac, A, Al, Z
${}^{98}Mo$	$3,5 \times 10^3$	CE, γ	0,3	Ac
${}^{100}Ru$	1	β^-	-	-
${}^{108}Ag$	130	CE, γ	0,4 - 0,6 - 0,7	Ac, A, Al
${}^{110}Ag$	0,7	β^-, γ	0,6 - 0,9	Ac, A, Al
137Cs	10,7	CE, γ	0,08 - 0,36	C
${}^{138}Cs$	2	γ	0,8	-
${}^{137}Cs$	30	β^-, γ	0,7	-
${}^{149}Ce$	0,7	β^-, γ	0,1	-
${}^{152}Sm$	93	β^-, γ	0,02	C
${}^{154}Eu$	13,4	β^-, γ, CE	0,1	C, G
${}^{158}Eu$	8,2	β^-, γ	0,1 - 1,3	C, G
${}^{223}U$	72	α, γ	0,05 - 1,3	-
${}^{225}U$	$1,6 \times 10^5$	α, γ	0,05 - 1	-
${}^{227}U$	$2,4 \times 10^5$	α, γ	0,05 - 0,1	-
${}^{228}U$	7×10^5	α, γ	0,2	-
${}^{227}U$	0,02	α, γ	0,2	-
${}^{228}I$	$4,5 \times 10^5$	β^-, γ	0,05	-
${}^{229}Np$	$2,1 \times 10^5$	α, γ	0,02 - 0,08	-
${}^{236}Pu$	87,7	α, γ	0,04 - 1,1	-
${}^{239}Pu$	$2,4 \times 10^4$	α, γ	0,05	-
${}^{240}Pu$	6537	α, γ	0,05 - 0,9	-
${}^{241}Pu$	1,7	α, γ	0,03 - 0,15	-
${}^{242}Pu$	$3,8 \times 10^5$	α, β^-, γ	0,05 - 0,09	-
${}^{243}Am$	432	α, γ	0,05 - 0,8	-
${}^{249}Am$	7380	α, γ	0,07 - 0,6	-

A: altri materiali; Ac: acciaio; Al: acciaio inox; Al: alluminio; C: calcestruzzo; CE: cattura elettronica; G: grafite; Z: leghe di zirconio.

Tab. 1. Principali radionuclidi rincontrati durante la caratterizzazione di impianti nucleari
Figure 18: Most common Radionuclides that are found during the Decommissioning of a NPP (Ref.[18])

All these radionuclides can be divided in three main different classes :

- **Major Neutron Activated Materials**

This part of the characterization is very important due to the fact that, as will be shown later, the neutron activated products represent the most important part of the plants radioactive inventories reaching up to hundreds of thousands of TBq for normal industrial reactors. Under will be provided a list of the major activation products with their formation mechanism, half life, decay characteristics and practical consequences of their presence.

- **TRITIUM (H3)** - Radionuclide that can be produced in a reactor by several mechanism. Neutron capture in Deuterium water (D2O) moderator is a major source of production in reactors using this technology. Also the concrete bio shield is a source of production with the $\text{Li}^6(n,\alpha)\text{H}^3$ reaction. Its half-life is 12.33y , and it decay with a β^- emission (maximum energy of about 19keV). (For what was said above is clear that tritium represent mainly a problem in CANDU type reactors, so the ones that use heavy water as a moderator; here large quantities of tritium are produced and easily transported all over the plant, creating non-negligible problems during the decommissioning);
- **CARBON (C14)** - This radionuclide (which also in very little part can be found in nature) is mainly produced by the activation of Nitrogen : $\text{N}^{14}(n,p)\text{C}^{14}$ (other less relevant activation reaction generating C14 are also the ones with C13). C14 has an half-life of 5730y , and it's a pure β^- emitter, with a maximum energy of 156keV. N14 can be found both in air and in construction materials, particularly in concrete and graphite. C14 is a major problem in reactors such as the CANDU, where nitrogen was sometimes used in the gap between the pressure tubes and the Calandria, and also for the RBMK reactors, where there's a relevant quantity of nitrogen cooled graphite moderator;
- **SODIUM (Na22 and Na24)** - For the sodium are listed two radionuclides : the Na22 is generated by fast neutrons reactions $\text{Na}^{23}(n,2n)\text{Na}^{22}$, and $\text{Na}^{23}(\gamma,n)\text{Na}^{22}$, it has an half life of 2.6y with two different type of decay possible, which are β^+ , γ , with a maximum energy of 1.275MeV to Ne22 . This radionuclides is cited because it will have to be taken in account in the future IV generation reactors decommissioning in future; the Na24 is on the other hand generated by neutron capture of the sodium impurities in H2O. It has an half-life of 15h , and it's a γ, β emitter with an energy that variates from 1.4 to 2.8 MeV;
- **CHLORINE (Cl36)** - Radionuclides principally produce by neutron capture by Cl35, or also from K39. It has an half life of 3.01×10^5 y with a β emission of maximum energy 709keV. It is mainly generated

by the presence of chlorine in most reactors construction materials, and it's important for the disposal due to its long half-life, and also due to its very high solubility in salts. One major source for its generation is for example the graphite in GCR's;

- **ARGON** (Ar39) - It's a radionuclide produced mainly by the reaction $K39(n,p)$ Ar39. It has a half-life of 269y , and it is a β^- emitter with a maximum energy of 565keV. Being K39 93% abundant in natural potassium, which is also present in materials such as concrete in concentrations of about thousands of ppm (or even in steel up to level of hundreds of ppm), Ar39 will become a significant volatile radionuclide some decades after the shutdown (This will happen specially in those reactors which use Argon as inert gas blanket, for ex. the fast breeder reactors);
- **CALCIUM** (Ca41) - Produced by the reaction $Ca40(n,\gamma)$ Ca41 , Ca41 then decays (with a half-life of 103000y) to K41 with an electronic capture related to a weak emission of X-rays. Calcium is one of the main component of concrete' bioshields, and it is also present in the graphite;
- **MANGANESE** (Mn54 and Mn56) - The first (Mn54) is formed with high energy neutron activation ($\gtrsim 10$ MeV) while the second one is formed with thermalized neutrons radiative capture. Mn54 has a half-life of 310d , while Mn56 of 2.6h . Activated Manganese derives from steel, and it could be also a corrosion product, but due to his short half-life its presence is relevant only for a short time after the shutdown;
- **IRON** (Fe55) - Produced by the reaction $Fe54(n,\gamma)$ Fe55 and decays with an half life of 2.73y by electronic capture with weak X-ray emission. It's very probable the translocation of Fe55 radionuclides to the coolant system, and being this a function of the corrosion, it will be a phenomenon more present in the carbon stainless steel than in other alloys steel;
- **NICHEL** (Ni59 and Ni63) - Ni59 is produced by neutron absorption by Ni58. It has a half-life of 76000y , and it decays by electronic capture with continuous X spectrum from inner bremsstrahlung, up to $\gtrsim 1$ MeV. It is very important for the waste disposal being the one considerate the limit for dose considerations after all the short lived had already decayed. On the other hand we have also Ni63, produced by neutron capture by Ni62. It has a half-life of 100.1y and it is a β^- emitter up to 67keV. Due to its very high presence in the alloys used for the reactors (inconel type) it is considered to be the most abundant activation product in a LWR;
- **COBALT** (Co60 and Co58) - The first isotope is produced by the activation with neutron absorption of the Co59 , it has a half-life of 5.27y with a maximum β^- emission of 318keV, and eventually decays

also γ producing up to 1.33MeV. The second one also is activated by neutron activation from Ni, and it has a lower half-life (71d) decaying with a maximum energy up to 811keV. Cobalt is both present in carbon and stainless steel, but being the major responsible dose producing radionuclide in the reactor interior from 10 to 50y , in NPP are used lower tenor of Cobalt alloy as possible it is;

- **ZINC** (Zn65) - It is produced by the reaction $Zn64(n,\gamma) Zn65$, then it decays (half-life of 244d) via electron capture or β^+ emission with energy up to 329eV. (It is present in units having Admiralty [29%Zn] or Munts[40%Zn] metal, which basically are the firsts BWR);
- **MOLYBDENUM** (Mo93) - Mo93 is produced with the neutron absorption of Mo92 , it decays by electronic capture to Nb93m (which then decays to Nb93) with low energies emitted (19keV), and it has a half-life of 3500y. It is present in concentrations up to 2 and 2.5% in certain stainless steels like SS316 and SS316L;
- **ZIRCONIUM** (Zr93) - It is produced by the reaction $Zr92(n,\gamma) Zr93$. Zirconium is present at about 98% in Zircaloy, which is the alloy used claddings for the fuels. It has a half-life of $1.5 \times 10^6 y$, and it decays β^- with an energy up to 60keV. After 1000y of decay Zr93 is considered the most important activation product in irradiated cladding/moderator tubes;
- **NIOBIUM** (Nb94) - It is produced with neutron capture from Nb93, it has a half-life of 20300y and it is a β^- emitter with a maximum energy up to 472keV to Mo94, which eventually decays γ more than one time. The high presence of Nb in stainless steel and inconel could lead to a significant amount of Nb94 in core materials, representing in this way the principal contributor to personnel exposure during the dismantle of reactor pressure vessel (On the other hand being very low soluble it is not affected in an important way by transport phenomena);
- **SILVER** (Ag108m and Ag110m) - Ag108m is produced by the reaction $107Ag(n,\gamma) Ag108m$, it has a half life of 130y and decays by weak electronic capture to 108Ag (which later with a β^- 1.655MeV emission decays to 108Cd). Ag110m is produced by the reaction $Ag109(n,\gamma) Ag110m$, it has a half-life of 249d and a β^- emission up to 1.467MeV; then Ag110m will decay (only after 24.5s) to stable 110Cd releasing with a β^- emission up to 2.893MeV. Ag radionuclides are present mainly in the control rods, due to the huge amount of silver into the PWR ones; they also are subject to corrosion and erosion, contaminating in this way the primary coolant (was also seen that the presence of Ag was related to the seals over the reactor vessel);
- **TIN/ANTIMONY** (Sn125/Sb125) - It is produced by the $Sn124(n,\gamma)$ Sn125 reaction , it has a half-life of 9.64d with a β^- decay of 2.35MeV

energy to Sb125, which after (a half life of 2.76y) decays β^- with an energy of 622keV (or by γ emission) to Te125m, which then decays to Te125 (Stable element) with a gamma emission. Sb125 can be found in Zircaloy irradiated cladding or moderator tubes, while Zn has a variability from 0.25% up to 1.7% depending on the zircaloy used;

- **BARIUM** (Ba133) - It is produced by the Ba132(n, γ) Ba133 reaction, it has a half life of 10.5y and decays with electronic capture or gamma emission to Cs133 stable isotopic composition. It is found mainly into the concrete (can reach up to the 40% by weight of it), where it is the main γ emitter;
- **CESIUM** (Cs134) - It is produced by the reaction Cs133(n, γ) Cs134 (which could also be a fission product). Cs134 has a half life of 2.065y and with a 658keV β^- decays to Ba134. Are also produced several gamma emissions during the decay;
- **EUROPIUM** (Eu152/154/155) - These radionuclides are produced both by neutron absorption of Eu151/153 or by chain absorption in Sm. Their half-life is respectively 13.5y, 8.6y, 4.76y, and they all decay β^- releasing 1.477MeV, 1.85MeV, and 2.52keV, to Gd with the same Eu atomic number (nb. While Gd154 and Gd155 are stable isotopes, Gd152 decays alpha). On a time scale of 10-20 years Europium 152/154 is the dominant activation product in bioshield concrete and core graphite and they have both very large neutron capture cross-section. Eu152 is produced by thermal neutrons mainly, while Eu154 has a substantial resonance integral. They have to be taken in consideration owing to the increasing presence of rare elements in the materials used for the parts cited above;
- **HOLMIUM** (Ho166m) - It's produced by the Ho165(n, γ) Ho166m reaction . It has a half-life of 1200y and it has a β^- decay of 1.314MeV. Ho165 is present in ppb quantities in steels, and in concrete with ppm quantities. It is characterized also by gamma rays, to which is the main contributor after 100y for graphite of GCRs.

In the next pages are reported some relevant data from Ref.[19].

Parent	Nuclear reaction	Daughter nuclide	Principal emissions	Half-life of daughter (a)	Abundance of parent nuclide in parent element (%)
Li-6	n, α	H-3	β^-	12.3	7.5
C-13	n, γ	C-14	β^-	5 730	1.1
N-14	n,p	C-14	β^-	5 730	99.6
Na-23	n,2n	Na-22	β^+ , EC	2.6	100
Na-23	γ ,n	Na-22	β^+ , EC	2.6	100
Cl-35	n, γ	Cl-36	β^- (β^+ , EC)	301 000	75.8
K-39	n,p	Ar-39	β^-	269	93.3
Ca-40	n, γ	Ca-41	EC	103 000	96.9
Fe-54	n,p	Mn-54	EC, γ	0.86	5.9
Mn-55	n,2n	Mn-54	EC, γ	0.86	100
Fe-54	n, γ	Fe-55	EC, X	2.7	5.9
Ni-58	n, γ	Ni-59	EC, X	76 000	68.3
Ni-62	n, γ	Ni-63	β^-	100	3.6
Co-59	n, γ	Co-60	β^- , γ	5.3	100
Zn-64	n, γ	Zn-65	EC, β^+	0.67	48.6
Zr-92	n, γ	Zr-93	β^-	1 500 000	17.1
Mo-92	n, γ	Mo-93	EC, X	3 500	14.8
Nb-93	n, γ	Nb-93m	IT, X	15.8	100
Nb-93	n, γ	Nb-94	β^- , γ	20 000	100
Mo-94	n,p	Nb-94	β^- , γ	20 000	9.3
Mo-98	n, γ	Tc-99	β^-	213 000	24.1
Ag-107	n, γ	Ag-108m	EC, γ	130	51.8
Ag-109	n, γ	Ag-110m	β^- , γ	0.68	48.2
Sn-124	n, γ	Sb-125	β^- , γ	2.76	5.8
Ba-132	n, γ	Ba-133	EC, X, γ	10.5	0.1
Eu-151	n, γ	Eu-152	EC, X, β^- , γ	13.5	47.8
Eu-153	n, γ	Eu-154	β^- , γ , X	8.6	52.2
Eu-154	n, γ	Eu-155	β^- , γ , X	4.76	0
Ho-165	n, γ	Ho-166m	β^- , γ , X	1 200	100

Figure 19: Most important activation reactions cited above (Ref.[19])

Also interesting could be watching the different main activation products in found in different type of reactors:

Radionuclides	Activity (Bq)	Radionuclides	Activity (Bq)
Fe-55	3.01E+15	H-3	3.7E+14
Co-60	1.89E+15	C-14	1.5E+13
Ni-63	7.26E+14	Cl-36	1.9E+11
Mn-54	2.81E+13	Ca-41	1.4E+11
Ni-59	5.97E+12	Fe-55	1.7E+16
H-3	3.73E+12	Ni-59	6.8E+12
Cs-134	4.63E+12	Co-60	3.6E+16
Ar-39	8.55E+12	Ni-63	7.4E+14
Ag-108m	4.20E+11	Mo-93	8.0E+11
Total	5.7E+15	Nb-94	1.4E+11
Assumptions: 870 MW(th), 23 years of irradiation, 10.6 EFPY, 5 years after shutdown.		Ag-108m	1.1E+11
		Sn-121m	4.5E+11
		Ba-133	8.4E+10
		Cs-134	5.9E+10
		Eu-152	3.1E+12
		Eu-154	4.2E+12
		Eu-155	1.9E+12
		Ho-166m	1.0E+10
		Tl-204	4.6E+13
		Total	5.4E+16
		Assumptions: 355 MW(th), 26 years of irradiation, 5 years after shutdown.	

(On the left the activation products found in **TRINO VERCELLSE PWR**, while on the right the ones found in a typical **UK MAGNOX**)

Radionuclides	Activity (Bq)	Radionuclides	Activity (Bq)
C-14	3.7E+10	Zr-95	1.28E+8
Ca-41	2.6E+8	Fe-55	1.45E+15
Mn-54	1.3E+10	Sb-125	7.00E+12
Fe-55	3.8E+13	Co-60	8.49E+14
Co-60	9.8E+13	H-3	5.49E+6
Ni-59	3.3E+11	Eu-152	2.25E+14
Ni-63	3.3E+13	Cd-113	7.72E+7
Nb-94	1.3E+9	Sn-121m	1.62E+11
Sb-125	4.8E+10	Ni-63	1.99E+14
Ba-133	3.8E+7	C-14	6.30E+11
Eu-152	1.7E+12	Nb-94	2.00E+12
Eu-154	1.7E+11	Ni-59	1.60E+12

Assumptions: 90 MW(th), 13 years of irradiation, 10 years after shutdown.

(On the left the data related to a **JPDR BWR** type, while on the right the data of a **GENTILLY-1 CANADA REACTOR STRUCTURE**)

^a Gentilly-1 is a boiling water, heavy water moderated reactor.

^b Structures include pressure tubes, calandria tubes, calandria side tube sheets, two sets of three axial shield slabs, outer tube sheets, reflector baffle and calandria, radial thermal shields (reactor vessel) and concrete biological shield.

^c Assumptions: 833 MW(th), 5 years of irradiation, 0.5 EFPY, 6 years after shutdown.

Figure 20: Different activation products depending on the reactor type (Ref.[19])

Here the data show us how the radiations are distributed in different components of different reactors type :

Components	Radioactivity (Bq)	Components	Radioactivity (Bq)
Internals	1.28E+16	Internals	4.27E+15
Fuel cases	2.44E+15	Control rods	1.16E+15
Control rods	3.4E+15	Vessel	3.52E+14
Reactor pressure vessel	3.84E+13	Neutron shield	2.47E+12
Sacrificial shield	6.18E+11	Biological shield	9.39E+9
Biological shield	3.33E+9		
Dry well	1.51E+10	Total	5.7E+15
Total	1.8E+16	Assumptions: 870 MW(th), 23 years of irradiation, 10.6 EFPY, 5 years after shutdown.	

Assumptions: 2590 MW(th), 7 years of irradiation, 4 effective full power years (EFPY), 5 years after shutdown.

Components	Radioactivity (Bq)	Components	Radioactivity (Bq)
Reactor pressure vessel (metal)	8.3E+15	Pressure vessel (without internals)	9.4E+12
Reactor internal structures (metal)	4.5E+16	Reactor pressure vessel lid	4.2E+7
Moderator (graphite)	4.4E+14	Lagging	8.0E+11
Reflector (graphite)	1.5E+14	Moderator tank	2.8E+16
Biological shield (concrete + steel reinforcing)	2.2E+13	Thermal shield	6.6E+14
Total	5.4E+16	Lower spacer	4.1E+14
Assumptions: 355 MW(th), 26 years of irradiation, 5 years after shutdown.		Upper spacer	6.8E+14
		Spacer ring	6.5E+13
		Fuel element channels	1.5E+14
		Control rod guide tubes	6.5E+13
		Total	3.0E+16

Assumptions: 200 MW(th), 18 years of irradiation, 10 EFPY (approx.), 5 years after shutdown.

(The data are respectively for: CAORSO BWR (top left corner), TRINO VERCELLESE PWR (top right corner), a typical UK MAGNOX GCR(bottom left corner), PHWR MZFR GERMANY TYPE(bottom right corner))

Figure 21: Distribution of activated materials in different reactors components (Ref.[19])

Before talking about the two remaining categories, the major fission products and the major actinides, we have to remark that these (that will be listed) are the radionuclides involved into the contamination of various materials. It could be loose or fixed contamination, and the removal method depends on the type of accident that happened. More in general these contaminants deposit on internal or external surfaces of the plant, and in majority they are related to the transport/leachout of fission products and actinides, creating a particular problem in those reactors characterised by a direct cycle to the turbine like BWRs or RBMKs. However the contamination can be found also in secondary cycle, and also in the storage pools, and whatever comes in contact with the fuel. It's also important to understand that the transport of contaminant is strictly related to the thermofluid dynamic of every single reactor, so it's not simple to individuate a common description of them changing from one reactor to another. In general the levels of radioactivity that these elements can lead to is in the order of the tens to hundreds of TBq. Fortunately the contamination is concentrated on the surfaces and not penetrate very deeply in the materials.

- **Major Fission Products**

Here follows a description of all the most relevant fission products radionuclides, with formation mechanism, half-life, decay characteristics and practical consequence of their presence, listed.

- **STRONTIUM (Sr90)** - Sr90 is principally produced by fission and its one of the most abundant fission products. Sr90 has a half-life of 28.7y and decays with β^- of 546keV to Y90, element with which strontium is in secular equilibrium. Y90 has a half-life of 64h and a β^- decay of 2.27MeV (very high). These are very dangerous radionuclide from the radio toxicity point of view (strontium is a alkaline earth metal which deposit directly in the bones, where with yttrium decays). It is present in the spent nuclear fuel roughly at the same quantity of Cs137;
- **KRIPTON (Kr85)** - This radionuclide has a half-life of 11y, it decays beta with a maximum energy of 690keV, or gamma with 510keV. It's a material with a low fission yield (0.3%). It's a noble gas, and due to the fact that it's inert it's more dangerous an eventual external radiation than an intake of it;
- **TECHNETIUM (Tc99)** - It can be a fission product with cumulative fission yields (is also produced with neutron capture and β^- decay of Mo98) , it has a half-life of 211100y , and with a 294keV β^- decays to Ru99. It's very dangerous if ingested;
- **RUTHENIUM (Ru106)** - This element produced by fission decays with 39keV β^- (half-life of 374d) into Rh106, that then will also decay (half-life 30s) with a 3.54MeV β^- to Pd106. Ruthenium can be present in different volatile species in high temperature conditions, and also deposit on metallic surfaces when soluted. Due to its very short life is not critical for the disposal.
- **IODIUM (I129 and I131)** - I131 is important only considering short times after the shut down of the reactor or considering accidental situations, and this to its very short half-life which is 8d. On the other hand is very dangerous because it has a medium fission yield (3%) and can volatilize easily, and in this way enters the human body, in particular fixing in the thyroid (which has a biological half life a lot grater than the physical half life of the element (120d) and in this way it all decays here, leading to cancer) and decaying (β^- of 610keV or gamma of 360keV). I129 has a half-life of 1.6×10^7 y and a 154keV β^- emission. Being produced by Te129 the quantity increases after irradiation, reaching a peak only after some months. It's not only long lived, but also a volatile radionuclide. Due to all this factors, if from the incidental point of view is more important the evaluation and protection from I131, from the waste disposal point of view is a lot more important the treatment of I129;

- **XENON** (Xe133 and Xe135) - Xenon radionuclides must be cited due to their importance during the operation of the plant (Xenon is in fact a neutron poisoner which must be strictly observed during the shut down of the reactor), but this radionuclides are not relevant from the disposal point of view because of their too short half-life, which are respectively 5.2d and 9h;
- **CESIUM** (Cs137) - Produced by fission, Cs137 has a half-life of 30y and decays with a maximum 1.17MeV β^- to Ba137 (around the 85% of decays are through Ba137m with the emission of 662keV photons). It's one of the most abundant fission products, with a fission yield of 6%. It is really soluble and due to this it's transported all around the LWR circuits. It's also a volatile isotope and it's due to this very dangerous for the decommissioning work. All the disposal facilities are mainly designed for this type of radionuclide;
- **CERIUM** (Ce144) - It has a half-life of 285d and decays with a 318keV β^- to Pr144, element with which is in secular equilibrium. Pr144 then (half-life of 17min) decays with 3MeV beta to a stable element. It has a high fission yield (like Cs around the 6%) but because of its very low half-life is not considered a critical radionuclide for the waste disposal.

• Major Actinides

The actinides that will be listed are mainly present into the spent nuclear fuel rods, but also can be found as contaminant in different materials in the plant due to leaching or previous accidents. Like was made above, will be listed for each radionuclide: formation mechanism, half-life, decay characteristics and practical consequence of their presence.

- **PLUTONIUM** (Pu238/239/241) - Plutonium can be both extracted from the fuel to be separated and reused as nuclear fuel, or directly treated as a waste and disposed. Pu isotopes can be produced by Np238 decay, or mainly by U235/238 neutron capture. They respectively have a half-life of 87.7y, 24110y, 14.35y, and they all have a α decay with high energy released (5.5MeV, 5.157MeV for 238 and 239 respectively) or a β low energy (21keV) for Pu241;
- **AMERICIUM** (Am241) - It's produced by the decay of Pu241, it has a half-life of 432y and a α emission of 5.486MeV , and in addition to this also a long term gamma emitter. Together with Pu is mainly found in crud accumulations and other corrosion products related to a fuel pin leakage. The presence of this long life alpha emitter radionuclides in materials, determine the disposing criteria that has to be used for different tools.
- **CURIUM** (Cm242 and Cm244) - The first is a member of uranium decay series, but it has a half life of 162.8d, so it's not so interesting for the disposal point of view, while on the other hand the second (a

member of thorium decay series) has a half-life of 18.1y, and it is a alpha emitter with an energy of 5.804MeV released;

- **URANIUM** - For what concern uranium, the characteristics of the isotopes which are more abundant in nature (232,235,238) were already discussed in the chapter where we talked about the NORM/TENORM waste. Here the differences are, for sure, the fact that we have much higher concentrations, and that other isotopes could be created, like U233, which is highly researched to be inserted into the thorium fuel cycle. Also, the problem related to these ones is mainly the fact that the reactions will give life to all the radionuclides listed until this point, but from the point of view of the disposal Uranium isotopes are not so important because they do not represent a waste, they are extracted mainly and reprocessed to be reused.

Under can be seen some tables where are listed the isotopic compositions of spent fuel from different NPP all over the world, estimated at different time after the shutdown, with different burnup levels and different enrichment to take a look of how the major actinides and the fission products can change their presence (This tables are all taken from Ref.[20], which for this analysis considered PWRs).

Assembly	NT3G23	NT3G23	NT3G23	NT3G23	NT3G23
Sample ID	SF95-1	SF95-2	SF95-3	SF95-4	SF95-5
Burnup ^a (GWd/MTU)	14.30	24.35	35.42	36.69	30.40
U-234	2.987E-04	2.850E-04	1.873E-04	1.870E-04	2.829E-04
U-235	2.674E-02	1.927E-02	1.326E-02	1.230E-02	1.544E-02
U-236	2.672E-03	4.024E-03	4.911E-03	4.999E-03	4.566E-03
U-238	9.499E-01	9.424E-01	9.338E-01	9.335E-01	9.388E-01
Pu-238	1.718E-05	7.102E-05	1.539E-04	1.588E-04	1.020E-04
Pu-239 ^b	4.227E-03	5.655E-03	6.194E-03	6.005E-03	5.635E-03
Pu-240	7.802E-04	1.539E-03	2.186E-03	2.207E-03	1.821E-03
Pu-241	3.690E-04	9.578E-04	1.486E-03	1.466E-03	1.153E-03
Pu-242	3.790E-05	1.844E-04	4.516E-04	4.803E-04	2.976E-04
Am-241	1.378E-05	2.344E-05	3.310E-05	2.351E-05	2.840E-05
Am-242m	1.840E-07	5.201E-07	7.877E-07	7.282E-07	5.687E-07
Am-243	2.682E-06	2.289E-05	8.047E-05	8.472E-05	4.400E-05
Cm-242	1.510E-06	7.672E-06	1.964E-05	2.328E-05	1.006E-05
Cm-243	1.415E-08	1.240E-07	3.720E-07	3.976E-07	2.293E-07
Cm-244	2.712E-07	5.042E-06	2.562E-05	2.837E-05	1.064E-05
Cm-245	5.519E-09	1.962E-07	1.396E-06	1.587E-06	4.839E-07
Cm-246	2.560E-10	1.190E-08	1.049E-07	1.251E-07	1.952E-08
Nd-142	3.429E-04	8.887E-06	2.116E-05	2.222E-05	1.371E-05
Nd-143	4.631E-04	7.149E-04	9.299E-04	9.373E-04	8.303E-04
Nd-144	3.276E-04	6.046E-04	9.347E-04	1.024E-03	7.928E-04
Nd-145	3.328E-04	5.384E-04	7.392E-04	7.598E-04	6.518E-04
Nd-146	2.809E-04	4.925E-04	7.340E-04	7.624E-04	6.185E-04
Nd-148	1.592E-04	2.736E-04	3.979E-04	4.126E-04	3.401E-04
Nd-150	7.200E-05	1.258E-04	1.895E-04	1.959E-04	1.572E-04
Cs-134	2.343E-05	7.012E-05	1.404E-04	1.471E-04	1.014E-04
Cs-137	5.405E-04	9.336E-04	1.347E-03	1.400E-03	1.148E-03
Ce-144	1.937E-04	3.160E-04	4.560E-04	4.301E-04	3.868E-04
Eu-154	4.093E-06	1.306E-05	2.525E-05	2.657E-05	1.817E-05
Ru-106	4.447E-05	8.340E-05	1.360E-04	1.401E-04	1.208E-04
Sb-125	1.671E-06	2.000E-06	2.707E-06	2.160E-06	2.667E-06
Assembly	NT3G23	NT3G23	NT3G23	NT3G23	NT3G23
Sample ID	SF96-1	SF96-2	SF96-3	SF96-4	SF96-5
Burnup ^a (GWd/MTU)	7.79	16.44	28.20	28.91	24.19
U-234	1.805E-04	1.522E-04	1.251E-04	1.250E-04	1.354E-04
U-235	1.944E-02	1.408E-02	8.638E-03	8.064E-03	9.937E-03
U-236	1.421E-03	2.411E-03	3.244E-03	3.302E-03	3.013E-03
U-238	9.660E-01	9.580E-01	9.476E-01	9.475E-01	9.522E-01
Pu-238	8.536E-06	4.172E-05	1.206E-04	1.248E-04	7.978E-05
Pu-239 ^b	3.781E-03	5.459E-03	6.001E-03	5.819E-03	5.519E-03
Pu-240	6.764E-04	1.494E-03	2.303E-03	2.327E-03	1.964E-03
Pu-241	2.622E-04	8.684E-04	1.498E-03	1.480E-03	1.203E-03
Pu-242	2.440E-05	1.615E-04	5.103E-04	5.411E-04	3.551E-04
Np-237	6.125E-05	1.323E-04	2.168E-04	2.252E-04	1.875E-04
Am-241	5.985E-06	1.735E-05	2.845E-05	3.094E-05	2.149E-05
Am-242m	1.218E-07	4.579E-07	6.413E-07	6.793E-07	5.647E-07
Am-243	1.147E-06	1.728E-05	8.872E-05	9.598E-05	5.078E-05
Cm-242	8.502E-07	5.781E-06	1.628E-05	1.679E-05	1.115E-05
Cm-244	9.560E-08	3.092E-06	2.862E-05	3.128E-05	1.280E-05
Nd-143	2.521E-04	4.778E-04	7.158E-04	7.184E-04	6.433E-04
Nd-144	1.536E-04	3.588E-04	7.292E-04	7.513E-04	5.927E-04
Nd-145	1.800E-04	3.575E-04	5.766E-04	5.880E-04	5.095E-04
Nd-146	1.536E-04	3.266E-04	5.795E-04	5.948E-04	4.910E-04
Nd-148	8.770E-05	1.851E-04	3.201E-04	3.280E-04	2.733E-04
Nd-150	4.130E-05	8.972E-05	1.591E-04	1.628E-04	1.331E-04
Cs-134	8.609E-06	3.759E-05	1.002E-04	1.047E-04	7.146E-05
Cs-137	2.813E-04	5.983E-04	1.018E-03	1.053E-03	8.572E-04
Ce-144	1.179E-04	2.250E-04	3.362E-04	3.453E-04	3.145E-04
Eu-154	2.309E-06	8.538E-06	1.973E-05	1.992E-05	1.423E-05
Ru-106	2.830E-05	6.053E-05	1.402E-04	1.291E-04	1.344E-04
Sb-125	1.433E-06	2.829E-06	3.658E-06	4.645E-06	3.690E-06

Figure 22: Takahama Unit 3 spent fuel samples (Ref.[20])

Nuclide	Measurement laboratory ^a	104-MKP109-LL (87-81)	104-MKP109-CC (87-72)	104-MKP109-P (87-63)
U-234	PNL	1.815E-01	1.588E-01	1.361E-01
U-235	PNL	9.609E+00	5.865E+00	4.016E+00
U-236	PNL	3.562E+00	4.005E+00	4.186E+00
U-238	PNL	9.558E+02	9.446E+02	9.358E+02
Pu-238	PNL	1.146E-01	2.147E-01	3.049E-01
Pu-239	PNL	4.837E+00	4.943E+00	4.943E+00
Pu-240	PNL	1.950E+00	2.540E+00	2.885E+00
Pu-241	PNL	7.725E-01	1.024E+00	1.157E+00
Pu-242	PNL	3.279E-01	6.535E-01	9.530E-01
Np-237	PNL	3.048E-01	4.048E-01	5.338E-01
Am-241	PNL	2.830E-01	3.901E-01	4.331E-01
Se-79	PNL	3.361E-03	4.459E-03	4.794E-03
Sr-90	PNL	3.784E-01	4.864E-01	5.425E-01
Tc-99	PNL	6.354E-01	8.149E-01	8.944E-01
Sn-126	PNL	1.149E-02	1.672E-02	2.022E-02
Cs-133	PNL	9.643E-01	1.237E+00	1.407E+00
Cs-135	PNL	4.096E-01	4.519E-01	4.874E-01
Cs-137	PNL	8.768E-01	1.177E+00	1.424E+00
Nd-143	PNL	6.954E-01	8.123E-01	8.656E-01
Nd-144	PNL	1.070E+00	1.518E+00	1.864E+00
Nd-145	PNL	5.786E-01	7.408E-01	8.440E-01
Nd-146	PNL	5.559E-01	7.737E-01	9.416E-01
Nd-148	PNL	3.006E-01	4.073E-01	4.855E-01
Nd-150	PNL	1.407E-01	1.951E-01	2.360E-01
Sm-147	KRI	2.303E-01	2.704E-01	3.081E-01
Sm-148	KRI	1.001E-01	1.612E-01	2.246E-01
Sm-149	KRI	2.893E-03 ^b	1.852E-03	4.558E-03
Sm-150	KRI	2.089E-01	2.896E-01	3.638E-01
Sm-151	KRI	7.521E-03	9.408E-03	1.072E-02
Sm-152	KRI	8.968E-02	1.096E-01	1.325E-01
Sm-154	KRI	3.066E-02	3.805E-02	5.640E-02
Eu-151	KRI	1.794E-03	1.045E-03	3.435E-03
Eu-152	KRI	1.157E-04	5.926E-05	4.473E-04
Eu-153	KRI	8.516E-02	1.290E-01	1.612E-01
Eu-154	KRI	6.075E-03	8.780E-03	1.175E-02
Eu-155	KRI	1.041E-03	1.385E-03	1.815E-03
Gd-154	KRI	1.365E-02	1.495E-02	2.002E-02
Gd-155	KRI	6.248E-03	7.408E-03	9.959E-03
Gd-156	KRI	6.364E-02	7.329E-02	9.533E-02
Gd-157	KRI		2.072E-03	4.887E-03
Gd-158	KRI	1.394E-02	1.486E-02	2.066E-02
Gd-160	KRI	1.446E-03		
Nuclide	Measurement laboratory ^b	106-NBD107-MM ^a	106-NBD107-GG (87-108)	106-NBD107-Q ^a
U-234	PNL	1.736E-01	1.441E-01	8.497E-02
U-235	PNL	4.379E+00	3.074E+00	1.595E+00
U-236	PNL	3.244E+00	3.437E+00	3.449E+00
U-238	PNL	9.581E+02	9.572E+02	9.384E+02
Pu-238	PNL	1.618E-01	2.209E-01	3.224E-01
Pu-239	PNL	4.327E+00	4.351E+00	4.272E+00
Pu-240	PNL	2.345E+00	2.633E+00	2.948E+00
Pu-241	PNL	8.236E-01	9.223E-01	1.005E+00
Pu-242	PNL	6.197E-01	8.795E-01	1.326E+00
Np-237	PNL	2.967E-01	3.645E-01	4.2905E-01
Am-241	PNL	3.901E-01	4.827E-01	7.207E-01
Se-79	PNL	3.088E-03	4.159E-03	4.425E-03
Sr-90	PNL	3.826E-01	4.271E-01	4.980E-01
Tc-99	PNL	5.101E-01	5.936E-01	7.221E-01
Rh-103	KRI		6.542E-01	
Sn-126	PNL	1.296E-02	1.470E-02	1.930E-02
Cs-133	PNL			
Cs-135	PNL	3.978E-01	4.086E-01	4.716E-01
Cs-137	PNL	9.761E-01	1.119E+00	1.464E+00
Nd-143	PNL		7.189E-01	
Nd-144	PNL		1.603E+00	
Nd-145	PNL		7.246E-01	
Nd-146	PNL		7.805E-01	
Nd-148	PNL		4.124E-01	
Nd-150	PNL		N/A	
Sm-147	KRI		2.659E-01	
Sm-148	KRI		1.713E-01	
Sm-149	KRI ^c		2.174E-03	
Sm-150	KRI		3.000E-01	
Sm-151	KRI		7.970E-03	
Sm-152	KRI		1.210E-01	
Sm-154	KRI		4.470E-02	
Eu-151	KRI		1.326E-03	
Eu-152	KRI		1.263E-04	
Eu-153	KRI		1.483E-01	
Eu-154	KRI		7.074E-03	
Eu-155	KRI		1.138E-03	
Gd-154	KRI		1.254E-02	
Gd-155	KRI		7.028E-03	
Gd-156	KRI		7.248E-02	
Gd-157	KRI		2.100E-03	
Gd-158	KRI		1.483E-02	
Gd-160	KRI		5.032E-04	

Figure 23: Calvert Cliffs Unit 1 spent fuel samples (Ref.[20])

Assembly	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU
Sample ID	A1B ^a	D2 ^b	B2 ^b	C1 ^b	D1A4 ^a	A2 ^b	C3 ^b	C2B ^a	B3J ^a	B1B ^a	D1A2 ^a
Burnup (GWd/MTU)											
U-234	44.8	44.8	50.1	50.2	50.5	50.6	51.3	52.6	53.0	54.5	55.7
U-235	2.054E-04	1.923E-04	1.868E-04	1.977E-04	1.975E-04	1.912E-04	1.847E-04	1.808E-04	1.834E-04	1.877E-04	1.927E-04
U-236	8.605E-03	7.378E-03	6.205E-03	6.588E-03	7.485E-03	6.319E-03	6.251E-03	6.228E-03	6.111E-03	6.386E-03	6.966E-03
Pu-238	5.111E-03	5.334E-03	5.400E-03	5.470E-03	5.362E-03	5.497E-03	5.327E-03	5.185E-03	5.456E-03	5.402E-03	5.452E-03
Pu-239	4.033E-04	3.252E-04	3.144E-04	3.299E-04	3.747E-04	3.538E-04	2.511E-04	4.585E-04	3.982E-04	4.316E-04	3.809E-04
Pu-240	5.064E-03	5.427E-03	5.289E-03	5.405E-03	5.399E-03	5.340E-03	5.512E-03	4.991E-03	5.088E-03	5.107E-03	5.452E-03
Pu-241	2.342E-03	2.667E-03	2.728E-03	2.754E-03	2.621E-03	2.781E-03	2.844E-03	2.546E-03	2.654E-03	2.632E-03	2.708E-03
Pu-242	1.208E-03	1.366E-03	1.387E-03	1.423E-03	1.430E-03	1.358E-03	1.403E-03	1.329E-03	1.364E-03	1.362E-03	1.469E-03
Pu-242	6.793E-04	7.945E-04	9.146E-04	9.000E-04	9.414E-04	9.229E-04	9.233E-04	9.318E-04	1.106E-03	9.570E-04	9.637E-04
Np-237	6.040E-04	6.755E-04	6.917E-04	7.041E-04	6.848E-04	6.938E-04	6.823E-04	6.864E-04	7.060E-04	7.012E-04	7.058E-04
Am-241	3.466E-04	3.457E-04	3.412E-04	3.770E-04	5.261E-04	3.021E-04	3.028E-04	5.074E-04	5.060E-04	2.880E-04	3.350E-04
Am-242m	--	--	--	--	8.389E-07	--	--	1.679E-06	1.244E-06	1.031E-06	6.085E-07
Am-243	1.245E-04	1.923E-04	2.552E-04	2.458E-04	1.846E-04	2.541E-04	2.465E-04	1.956E-04	2.111E-04	2.043E-04	2.056E-04
Nd-143	9.850E-04	9.134E-04	9.987E-04	9.794E-04	1.080E-03	9.516E-04	9.510E-04	1.033E-03	1.060E-03	1.086E-03	1.111E-03
Nd-145	8.521E-04	8.289E-04	9.062E-04	8.972E-04	9.598E-04	8.777E-04	8.965E-04	9.411E-04	9.770E-04	9.847E-04	1.000E-03
Nd-148	4.869E-04	4.869E-04	5.447E-04	5.452E-04	5.482E-04	5.506E-04	5.577E-04	5.720E-04	5.761E-04	5.926E-04	6.058E-04
Cs-137	1.682E-03	1.617E-03	1.748E-03	1.811E-03	1.652E-03	1.765E-03	1.699E-03	1.762E-03	1.733E-03	1.758E-03	1.533E-03
Sm-147	2.258E-04	1.821E-04	1.859E-04	1.866E-04	2.353E-04	1.968E-04	1.819E-04	2.288E-04	2.479E-04	2.549E-04	2.515E-04
Sm-149	3.113E-06	3.094E-06	3.264E-06	3.188E-06	3.599E-06	3.816E-06	2.899E-06	3.358E-06	3.189E-06	3.423E-06	3.855E-06
Sm-150	3.577E-04	3.485E-04	3.754E-04	3.835E-04	4.125E-04	3.742E-04	3.619E-04	4.189E-04	4.525E-04	4.675E-04	4.525E-04
Sm-151	1.292E-05	1.264E-05	1.341E-05	1.247E-05	1.412E-05	1.256E-05	1.256E-05	1.329E-05	1.475E-05	1.500E-05	1.551E-05
Sm-152	1.217E-04	1.208E-04	1.295E-04	1.266E-04	1.338E-04	1.321E-04	1.256E-04	1.301E-04	1.419E-04	1.436E-04	1.423E-04
Eu-151	6.579E-07	7.034E-07	7.934E-07	6.856E-07	6.673E-07	8.832E-07	8.476E-07	7.030E-07	7.475E-07	5.696E-07	6.618E-07
Eu-153	1.468E-04	1.561E-04	1.674E-04	1.672E-04	1.744E-04	1.709E-04	1.607E-04	1.725E-04	1.834E-04	1.859E-04	1.891E-04
Eu-155	1.004E-05	1.227E-05	1.313E-05	1.432E-05	1.264E-05	1.284E-05	1.274E-05	9.964E-06	1.032E-05	1.546E-05	9.821E-06
Assembly	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU	NJ05YU
Sample ID	A1B ^a	D2 ^b	B2 ^b	C1 ^b	D1A4 ^a	A2 ^b	C3 ^b	C2B ^a	B3J ^a	B1B ^a	D1A2 ^a
Burnup (GWd/MTU)											
Gd-155	8.224E-06	5.594E-06	6.547E-06	6.357E-06	1.394E-05	5.220E-06	6.666E-06	9.411E-06	1.041E-05	1.003E-05	1.019E-05
Mo-95	1.041E-03	9.199E-04	1.128E-03	1.100E-03	1.089E-03	1.118E-03	1.006E-03	1.098E-03	1.124E-03	1.150E-03	1.111E-03
Tc-99	1.422E-03	9.757E-04	1.091E-03	1.081E-03	1.191E-03	1.081E-03	1.034E-03	1.356E-03	1.244E-03	1.316E-03	1.138E-03
Ru-101	1.115E-03	9.478E-04	1.202E-03	1.164E-03	1.098E-03	1.155E-03	1.025E-03	1.172E-03	1.171E-03	1.187E-03	1.129E-03
Rh-103	5.956E-04	5.157E-04	6.288E-04	6.182E-04	6.027E-04	6.190E-04	5.475E-04	6.145E-04	6.203E-04	6.267E-04	6.168E-04
Ag-109	5.111E-05	4.655E-05	5.280E-05	5.359E-05	8.463E-05	5.968E-05	9.233E-05	6.532E-05	7.788E-05	4.399E-05	4.607E-05

Figure 24: TMI Unit 1 spent fuel samples (Ref.[20])

1.4.2 Radioactive Sources

As anticipated in the previous part, most of the HLW come from NPP. Analysing these, is also possible to observe that the majority of them, even the most dangerous from the radioprotection point of view, are produced during the fuel cycle.

Nuclear reactor fuel contains ceramic pellets of uranium-235 inside of metal rods. Before these fuel rods are used, they are only slightly radioactive and may be handled without special shielding. During the fission process, two things happen to the uranium in the fuel. First, uranium atoms split, creating energy that is used to produce electricity. The fission creates radioactive isotopes of lighter elements such as cesium-137 and strontium-90. These isotopes, called "fission products," account for most of the heat and penetrating radiation in high-level waste. Second, some uranium atoms capture neutrons produced during fission. These atoms form heavier elements such as plutonium. These heavier-than-uranium, or "transuranic," elements do not produce nearly the amount of heat or penetrating radiation that fission products do, but they take much longer

to decay. Transuranic wastes, sometimes called TRU, account for most of the radioactive hazard remaining in high-level waste after 1,000 years. Radioactive isotopes eventually decay, or disintegrate, to harmless materials. Some isotopes decay in hours or even minutes, but others decay very slowly. Strontium-90 and cesium-137 have half-lives of about 30 years (half the radioactivity will decay in 30 years). Plutonium-239 has a half-life of 24,000 years. High-level wastes are hazardous because they produce fatal radiation doses during short periods of direct exposure. For example, 10 years after removal from a reactor, the surface dose rate for a typical spent fuel assembly exceeds 10,000 rem/hour – far greater than the fatal whole-body dose for humans of about 500 rem received all at once. If isotopes from these high-level wastes get into groundwater or rivers, they may enter food chains. The dose produced through this indirect exposure would be much smaller than a direct-exposure dose, but a much larger population could be exposed. Reprocessing separates residual uranium and plutonium from the fission products. The uranium and plutonium can be used again as fuel. Most of the high-level waste (other than spent fuel) generated over the last 35 years has come from reprocessing fuel from government-owned plutonium production reactors and from naval, research and test reactors. A small amount of liquid high-level waste was generated from reprocessing commercial power reactor fuel in the 1960s and early 1970s.

(*Obs. Even in the U.S. there is no commercial reprocessing of nuclear power fuel at present.*)

Almost all existing commercial high-level waste is unprocessed spent fuel.

All these HLW originated in the fuel cycle, can be divided into two main categories :

- **Front End :**

Waste from the front end of the nuclear fuel cycle is usually alpha-emitting waste from the extraction of uranium. It often contains radium and its decay products. Uranium dioxide (UO_2) concentrate from mining is a thousand or so times as radioactive as the granite used in buildings. It is refined from yellowcake (U_3O_8), then converted to uranium hexafluoride gas (UF_6). As a gas, it undergoes enrichment to increase the U-235 content from 0.7% the U-238 isotope, with a U-235 content of 0.3%

- **Back End :**

The back-end of the nuclear fuel cycle, mostly spent fuel rods, contains fission products that emit beta and gamma radiation, and actinides that emit alpha particles, such as uranium-234 (half-life 245 thousand years), neptunium-237 (2.144 million years), plutonium-238 (87.7 years) and americium-241 (432 years), and even sometimes some neutron emitters such as californium (half-life of 898 years for californium-251). These isotopes are formed in nuclear reactors. It is important to distinguish the processing of uranium to make fuel from the reprocessing of used fuel. Used fuel contains the highly radioactive products of fission (see high-level waste below). Many of these are neutron absorbers, called neutron poisons in this context. These eventually build up to a level where they absorb so

many neutrons that the chain reaction stops, even with the control rods completely removed. At that point, the fuel has to be replaced in the reactor with fresh fuel, even though there is still a substantial quantity of uranium-235 and plutonium present. In the United States, this used fuel is usually "stored", while in other countries such as Russia, the United Kingdom, France, Japan, and India, the fuel is reprocessed to remove the fission products, and the fuel can then be re-used. The fission products removed from the fuel are a concentrated form of high-level waste as are the chemicals used in the process. While most countries reprocess the fuel carrying out single plutonium cycles, India is planning multiple plutonium recycling schemes and Russia pursues closed cycle.

Then are present also other types of radioactive waste, which depending on their half-life and activity, can be classified either in HLW, ILW, or LLW, and then disposed. These are materials which have been activated or contaminated during the operational time :

- **Neutron activated materials :**

These materials are located in and near the core and have been irradiated by neutrons. The reactor core is the most activated part of the reactor structure. The portion of the reactor exposed to relatively low neutron fluxes is essentially the biological shield, usually made of concrete and steel reinforcements;

- **Contaminated materials :**

Contamination arises from the activation of the corrosion and erosion products conveyed by the coolant and from the dispersion of the irradiated fuel and fission products through cladding breaches. In addition, contamination results from leakages in the primary circuit, processing and storage of radioactive effluents and wastes, maintenance and repair activities, fuel discharging operations and working incidents. Airborne contamination may also give rise to a deposit of radioactive substances on walls, ceilings and in the ventilation system. It is assumed that the nuclear fuel and process fluids have been removed from the reactor after shutdown, before any decommissioning work. However, in some cases, especially where the reactor has undergone abnormal operational conditions such as major fuel element failure, residues of these materials will remain and must be included in the inventory.

Under, a list of all the materials/tools which are either activated or contaminated and that need to be disposed.

Radioactive waste from research reactors

- Gaseous Radioactive Waste

The typical sources of gaseous radioactive waste generated in the operation of a research reactor include the following:

- * Gaseous radioactive elements or compounds from the reactor pool, the coolant system, irradiation facilities and experimental facilities;
- * Airborne radioactive material produced in ancillary facilities, including fume cupboards and decontamination areas.

– Liquid Radioactive Waste

The typical sources of liquid radioactive waste generated in the operation of a research reactor include the following:

- * Cooling water draw-off;
- * Primary system drains (in the case of light water reactors);
- * Liquid waste from the demineralized water system;
- * Runoff from the drain of the ventilation water system;
- * Demineralized wastewater recovered from the drainage of large items of equipment in maintenance operations;
- * Runoff from washbasins and showers;
- * Runoff from floor drains;

– Solid Radioactive Waste

The typical sources of solid radioactive waste generated in the operation of a research reactor include the following:

- * Irradiated target cans;
- * Used irradiation rigs and reactor components (e.g. thermocouples);
- * Neutron beam guide tubes;
- * Spent ion exchange resins;
- * Used control rods;
- * Waste arising from the pool service area;
- * Waste from ventilation systems (charcoal filters, high efficiency particulate air (HEPA) filters);
- * Cleaning materials and used personal protective items;
- * Laboratory waste (gloves, tissue paper, disposable glassware, etc.);
- * Contaminated items arising from maintenance and other works.

Radioactive waste from nuclear power plants

– Gaseous Radioactive Waste

Depending on the type of nuclear power plant, possible sources of gaseous radioactive waste include the following

- * Leakage from the coolant;
- * The moderator systems of the reactor itself;
- * Degasification systems for the coolant;

- * Condenser vacuum air ejectors or pumps;
- * The exhaust from turbine gland seal systems;
- * Activated or contaminated ventilated air.

For all types of nuclear power plant, spent fuel in storage or in handling operations is a potential source of gaseous radioactive waste.

- Liquid Radioactive Waste

The primary coolant in water cooled reactors and water from the fuel storage pools are major sources of liquid radioactive waste, as some of their radioactive content may be transported to the liquid radioactive waste stream via process streams or leakages. Although the composition of liquid radioactive waste may differ depending on the type of nuclear power plant, contributions to the liquid waste stream may derive from the following

- * Reactor coolant letdown;
- * Evaporator concentrates;
- * Runoff from equipment drains;
- * Runoff from floor drains;
- * Laundry waste;
- * Contaminated oil;
- * Waste arising from the decontamination and maintenance of facilities and equipment.

- Solid Radioactive Waste

Solid radioactive waste is generated in the operation, maintenance and decommissioning of a nuclear power plant and its associated processing systems for gaseous and liquid radioactive waste. The nature of such waste varies considerably from plant to plant, as do the associated levels of activity. Solid radioactive waste may consist of the following

- * Spent ion exchange resins (both bead resins and powder resin);
- * Cartridge filters and pre-coat filter cake;
- * Particulate filters from ventilation systems;
- * Charcoal beds;
- * Tools;
- * Contaminated metal scrap;
- * Core components;
- * Debris from fuel assemblies or in-reactor components;
- * Contaminated rags, clothing, paper and plastic.

1.4.3 Conclusions

1.5 General Conclusions

1.6 References

1. INTERNATIONAL ATOMIC ENERGY AGENCY, Classification of Radioactive Waste, General Safety Guides, 2009;
2. INTERNATIONAL ATOMIC ENERGY AGENCY, Application of the Concepts of Exclusion, Exemption and Clearance, IAEA Safety Standards Series No. RS-G-1.7, IAEA, Vienna (2004);
3. ‘An Introduction to Nuclear Waste Immobilization’;
4. EUROPEAN COMMISSION, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, General Safety Requirements, 2014;
5. Decreto 7 agosto 2015 “Classificazione dei rifiuti radioattivi, ai sensi dell’articolo 5 del decreto legislativo 4 marzo 2014, n. 45” (GU 19 agosto 2015, Serie generale n. 191);
6. Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) | US EPA;
7. ‘An Introduction to Nuclear waste Immobilization’, chapter-5;
8. Evaluation of Guidelines for Exposures to Technologically Enhanced Naturally Occurring Radioactive Materials (<http://www.nap.edu/catalog/6360.html>);
9. Professor Campi Radioprotection Course material;
10. https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/946164/JSP_392_Chapter_29_-_THORIUM_Alt_Text_.pdf;
11. NuDat online database maintained by Brookhaven National Laboratory, September 9, 1997;

12. Report to Congress : evaluation of EPA's guidelines for Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) (pdf).
13. Naturally Occurring Radioactive Materials in Coals and Coal Combustion Residuals in the United States (2015);
14. EPA - A Regulators' Guide to the Management of Radioactive Residuals from Drinking Water Treatment Technologies;
15. ISCORS Assessment of Radioactivity in Sewage Sludge: Radiological Survey Results and Analysis;
16. IAEA-TECDOC-1183 "Management of radioactive waste from the use of radionuclides in medicine" - IAEA, November 2000;
17. Waste Technology Section International Atomic Energy Agency Wagramer Strasse 5 P.O. Box 100 A-1400 Vienna, Austria;
18. 'Decommissioning degli impianti nucleari e gestione dei rifiuti radioattivi' , Giambattista Guidi ;
19. 'Radiological characterization of shut down nuclear reactors for decommissioning purposes' , International Atomic Energy Agency, Vienna, 1998;
20. Predictions of PWR Spent Nuclear Fuel Isotopic Composition, March 2010, G.Radulescu I.C.Gauld G.Ilas;
21. IAEA "Predisposal Management of Radioactive Waste from Nuclear Power Plants and Research Reactors" ;
22. U.S.NRC website: "backgrounder on Radioactive Waste";