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RADIOACTIVE RISK SET



Volume 5

Management of Radioactive Waste

Jean-Claude Amiard

ISTE

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coordinated by
Jean-Claude Amiard

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Preface

The use of nuclear energy for military or civilian purposes inevitably leads to the production of radioactive waste. The management of this waste is one of the most serious problems facing industrialized nations.

As with all other wastes, radioactive waste can be disposed of in one of two ways: dilution or containment. A third method exists for radioactive waste with a very short physical life, less than 100 days, which is to wait, under safe conditions, for natural physical decay.

Dilution consists of reducing the radioactive risk by dispersing the radionuclides in vast compartments of the environment such as the lithosphere, the atmosphere or the hydrosphere. This can only be done for very low-level radioactive waste, even though it has been practiced more widely in the past.

Containment consists of immobilizing the waste as long as it remains radioactive. This is relatively easy for short-lived radionuclides, i.e. with a physical half-life of less than 30 years. On the contrary, it is much more difficult to ensure for long-lived radionuclides, for some of which the physical half-life is counted in millions of years. Currently, the only realistic and practicable solution found is the multiplication of physical barriers between the radioactive waste and the environment and the biosphere, the last barrier being geologically stable and impermeable layers of the lithosphere.

The classification of radioactive waste has been the subject of IAEA recommendations, but this has not prevented the multiplication of classifications in different states, which complicates possible comparisons. These classifications are based on a combination of two parameters: the waste's level of activity and the half-life of the radionuclides constituting the waste.

A major difference in classification divides nations into two categories depending on whether they practice an open or closed nuclear fuel cycle. In the latter case, a portion of the radioactive waste is removed from this classification and is considered as usable nuclear material. However, the number of states using the closed cycle is steadily decreasing, which makes it necessary to review the quantities of radioactive waste to be actually managed.

The management of radioactive waste is specific to each state. The majority of nations manage short-lived radioactive waste in surface storage facilities and a minority in underground facilities.

On the contrary, for long-lived radioactive waste, few states have definitive solutions. This is due to the fact that the containment of the radionuclide must be guaranteed for thousands of years. For low-level waste, most countries opt for dry interim storage. For intermediate- and high-level waste, the solution generally envisaged is deep geological disposal, with some countries favoring deep geological drilling.

In the field of radioactive waste management, research is very active and innovations are numerous. This does not prevent gaps in our knowledge, uncertainties about the nature of the disposal to be adopted for certain categories of waste and often a negative opinion of the public to the proposed solutions.

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Classifications and Origins of Radioactive Waste

1.1. Introduction

Compared to other categories of waste, the quantity of radioactive waste is relatively small. In France, nuclear waste represents 2 kg per year per inhabitant [AND 17a], compared to 580 kg of household waste, 900 kg of non-construction waste and 3.4 tons of industrial waste [ADE 20]. But these residues represent an immense problem because some of them are extremely radioactive and remain harmful over excessively long time scales, for some hundreds of thousands or millions of years, that humanity cannot control.

What can we do with this radioactive waste? In the past, the ocean has served as a dumping ground for nuclear powers, which have immersed tens of thousands of radioactive drums. This time is fortunately over. Some eccentric people have suggested dropping them into space. Fortunately, the idea was not pursued. The solution now being considered for the most dangerous waste is to bury it in deep layers of clay, granite, salt or tuff, hoping that nature and geology will compensate for the weaknesses of human technology [AMI 13]. Sweden was the first nation to choose an underground storage site. All other countries, faced with the concerns of their populations and the vagaries of political changes, have postponed their decisions. On the contrary, in the United States, the suspension of the Yucca Mountain storage project in Nevada, which was ready to open, is a sign of the American administration's desire to listen to the public. However, the State must find a new solution.

Since no alternative solution is yet mature, we must take our time in making a decision that will commit humanity for a long time. France, like Canada, Switzerland and Japan, has made the principle of reversibility central to its doctrine. On the contrary, Sweden and Finland do not require it, and the United Kingdom is still

considering it. It is not only a question of being able to recover radioactive packages, but of leaving the decision-making process open and giving it back to the political institutions. Parliament has once again become the master of nuclear waste management and future generations have the guarantee that nothing will be decided inescapably. The approach is virtuous. Let us hope that it is not an admission of powerlessness in the face of an insoluble puzzle [AMI 13]. It should also be emphasized that this postponement amounts in practice to leaving to future generations the care to manage and pay for the waste produced by the present generation.

Those responsible for the civilian and especially the military use of nuclear energy have in the past been very unaware of the seriousness with which the problem of nuclear waste is treated today. For example, the Hanford site in the United States was heavily polluted by unauthorized dumping during intensive plutonium production after World War II. Recently, six underground tanks leaked. In the former Soviet Union (USSR), waste in the form of highly active liquid solutions was injected directly into deep storage [MAC 96]. The United Kingdom in particular, but also other countries, and even France, have thrown drums of waste into international waters, a practice that is now prohibited [CAS 02].

Nuclear energy has been questioned almost since its inception and one of the main problems concerning its social acceptability in the world is the management of nuclear waste [ROD 17]. It is therefore imperative that nuclear nations manage radioactive waste in an exemplary way.

1.2. What is radioactive waste?

A few definitions should be kept in mind. Radioactive waste is radioactive material for which no further use is planned or envisaged. Ultimately radioactive waste is radioactive waste that can no longer be treated under current technical and economic conditions, in particular by extracting its recoverable part or by reducing its polluting or dangerous nature (French Environmental Code, article L 542.1-1). Conversely, if a radioactive material also contains radionuclides, it has a potential future use. This is the case for depleted uranium or spent nuclear fuel that can eventually be reused.

A radioactive substance is a substance that contains radionuclides, natural or artificial, whose activity or concentration justifies radiation protection control. The radionuclides contained in radioactive waste can be of artificial origin, such as cesium-137, or natural origin, such as radium-226.

Radioactive waste has three main characteristics, the type of radionuclide, the activity and the half-life. The type of radionuclide contained is related to the

radiation emitted (alpha, beta, gamma). The activity is the number of atomic nuclei that spontaneously disintegrate per unit of time; it is expressed in becquerels (Bq). The half-life is the time required for the activity of a radionuclide in a sample to decrease by half [IRS 13a, IRS 13b].

1.3. Classifications of nuclear waste

Waste classification is not unique. Indeed, while the IAEA has provided broad guidelines for defining and classifying radioactive waste, each state is free to use its own nomenclature.

1.3.1. General information on the classification of radioactive waste

As regards the classification of radioactive waste, there are two main approaches: one by a waste management channel and the other by a waste production channel. The latter approach is partly inherited from the historical concept of radiation protection.

The management pathway approach often combines the activity and lifetime parameters of the radionuclides constituting the waste. This classification was recommended by the IAEA in the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management. This classification is used in France, Belgium and Spain. Sometimes this approach is based only on activity. In Canada, for example, there are only three main categories of radioactive waste (ILW, HLW and spent fuel), except for the specific management of waste from mines. In the Netherlands, the classification has a larger number of categories, but no distinction is made between short- and long-lived waste and consequently there are no plans for surface disposal. In Germany, the classification is based mainly on the exothermic character of the waste.

The production chain approach leads to a more complex classification, with specific chains for certain types of waste, and combining activity and lifespan. This is the approach of the United States, Japan and Sweden (in fact in Sweden, the two types of approach coexist). In Finland, a category is sometimes added for waste from hospitals, universities, etc.

There are also national specificities, as in Belgium, which treats 50% of the radium sources used in the world (the result of uranium mining in the Congo, which is historically Belgian), or in Canada, which has large uranium mines. Similarly, in France, it should be noted that there is no release threshold for waste containing, or likely to contain, only very small quantities of radioactive elements [AMI 13].

1.3.2. The IAEA's recommendations

The IAEA proposes dividing radioactive waste into five categories, in addition to the category of waste considered as released (EW, Exempt Waste), according to two criteria, the amount of activity and the half-life of the radionuclide (Figure 1.1). These categories are very short-lived waste (VSLW), very low-level waste (VLLW), low-level waste (LLW), intermediate-level waste (ILW) and high-level waste (HLW) [IAE 09a].

In certain circumstances, such as acceptance into a radioactive waste disposal facility, Waste Acceptance Criteria (WACs) may be established for certain radionuclides. WACs are quantitative or qualitative criteria that may include, for example, restrictions on the activity concentration or total activity of particular radionuclides (or types of radionuclides) in the waste, or requirements regarding the form or packaging of the waste.

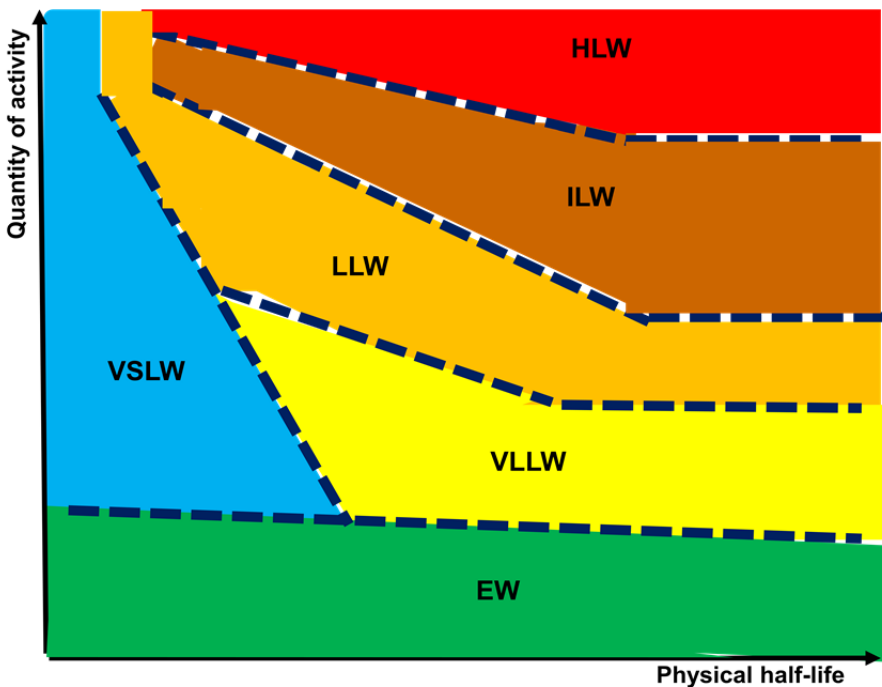


Figure 1.1. Proposed IAEA classification of radioactive waste (source: [IAE 09a]). EW: exempt waste; HLW: high-level waste; ILW: intermediate-level waste; LLW: low-level waste; VLLW: very low-level waste; VSLW: very short-lived waste. For a color version of this figure, see www.iste.co.uk/amiard/radioactive.zip

1.3.3. The French classification of radioactive waste

The details of the French classification are as follows. Radioactive waste is classified according to two criteria: mass activity and physical half-life. The “mass activity” criterion divides waste into four groups: *déchets de très faible activité*, called TFA or very low-level waste (VLLW), *déchets de faible activité*, FA or low-level waste (LLW), *déchets de moyenne activité*, MA or intermediate-level waste (ILW) and *déchets de haute activité*, high-level waste (HLW). The “life” criterion is divided into three classes to distinguish between *déchets à vie courte*, short-lived waste (SLW), *déchets à vie moyenne*, medium-lived waste (MLL) and *déchets à vie longue*, long-lived waste (LLW). The combination of the two criteria makes it possible to classify the waste into 12 categories (Table 1.1) [PNG 10].

	Very short life (VSL) <100 days	Short life (SL) ≤31 years old	Long life (LL) More than 31 years old
Very low activity (VLL) <100 Bq.g ⁻¹	VLL-SL Morvilliers	VLL-SL Aube Center	VLL-LL Aube Center
Low activity (LLW) Thousands of Bq.g ⁻¹	LLW-VSL Beaumont-Hague, Soulaines	LLW-SL Aube Center	LLW-LL (call for applications)
Average activity (AA) Millions of Bq.g ⁻¹	AA-VSL Beaumont-Hague, Soulaines	AA-SL Aube Center	AA-LL Bure?
High activity (HLW) Billions of Bq.g ⁻¹	HLW-VSL Not applicable	HLW-SL Bure?	HLW-LL Bure?

Table 1.1. French classification of radioactive waste and storage sites in operation in France (source: modified from [PNG 10, MTE 18]). For a color version of this table, see www.iste.co.uk/amiard/radioactive.zip

Radioactive waste management simplifies these subdivisions by grouping certain categories to manage them together. In the end, in France, by combining the four levels of activity with the three ranges of radioactive periods, six categories of waste are distinguished, defined by an order of April 4, 2014. In addition, this decree defines the nature of the information that nuclear activity managers and companies are required to establish, maintain and periodically transmit to ANDRA.

At present, only two categories have well-defined channels: VLA-SL at Morvilliers and LA-SL and AA-SL at Soulaines in the Aube region (and previously in the commune of La Hague, at the *Centre de stockage de la Manche-CSM*, 1969–1994). The other channels are still being studied, as are certain specific wastes such as tritiated waste, mining waste, sealed sources and graphite waste (see Chapter 5).

1.3.3.1. Activity levels used in France

Based on their activity levels, nuclear waste can be classified into the following six categories:

- Very short-lived waste (VSL) is managed by allowing it to decay on site and then it is disposed of in conventional channels. It is therefore not sent to a storage facility dedicated to radioactive waste.

- Very low-level waste (VLLW) comes from the operation of nuclear power plants and research centers, from fuel cycle facilities and research centers. The activity level of this waste is generally less than 100 Bq.g^{-1} . However, the management of this waste justifies radiation protection monitoring.

- Low-level and intermediate-level short-lived waste (LL/IL-SLW) come from the operation and dismantling of nuclear power plants and research centers and, for a small part, from biomedical research activities. The activity of this waste is between a few hundred Bq.g^{-1} and 1 million Bq.g^{-1} .

- Long-lived low-level waste (LL-LLW) consists mainly of graphite waste and radium-bearing waste. Graphite waste has an activity of between 10,000 and 100,000 Bq.g^{-1} , essentially long-lived beta emitting radionuclides. It comes from the dismantling of first-generation nuclear power plants (UNGG). Radium-bearing waste, mostly from non-nuclear industrial activities, is mainly composed of long-lived alpha-emitting radionuclides and has an activity of between a few tens of Bq.g^{-1} and a few thousand Bq.g^{-1} .

- Long-lived intermediate-level waste (LL-ILW) comes mainly from spent fuel reprocessing activities. It is technological waste (used tools, equipment, etc.), waste from the treatment of effluents such as bituminous sludge and structural waste, the shells and end caps that make up the nuclear fuel cladding, packaged in cemented or compacted waste packages. The activity of this waste is of the order of 1 million to 1 billion Bq.g^{-1} .

- High-level waste (HLW) also consists mainly of vitrified waste packages from the reprocessing of spent fuel. These waste packages concentrate the great majority of radionuclides, whether fission products or minor actinides. The activity level of this waste is of the order of several billion Bq.g^{-1} [JOR 14].

1.3.3.2. *French radioactive waste systems*

As Table 1.1 indicates, not all categories of waste have their storage site yet closed in France. We will detail this aspect later (Chapter 5).

Two important aspects condition the classification of radioactive waste. The first aspect is that there is no single classification criterion for determining a waste class. It is indeed necessary to study the activity of the different radionuclides present in the waste to position it in the classification. However, in the absence of a single criterion, the wastes in each category generally fall within a range of mass activity indicated below.

The second aspect is that a particular type of waste may fall into a defined category but not be accepted in the corresponding management channel because of other characteristics (e.g. its chemical composition or physical nature, such as radium-bearing waste that emits a radioactive gas, radon-222). Consequently, the waste category is not necessarily assimilated to its management channel [AMI 13].

1.3.3.3. *Hospital radioactive waste*

With respect to hospital radioactive effluents, French legislation is very strict and requires the intervention of official institutions, in particular ANDRA, for the conditioning, elimination, transport and storage of this waste [FRE 01, ACR 12]. This statement must be moderated, however, in view of the increase in practices involving radionuclides. The next radionuclides to be used will be beta and especially alpha emitters, which have a limited range in living matter. Recently, research is therefore exploring a number of products under development using isotopes such as lutetium-177, promethium-149, bismuth-212, bismuth-213, astatine-211, radium-223 and polonium-210.

1.3.3.4. *Harmfulness of radioactive waste*

For France, the IRSN [IRS 18b] proposes a methodology and possible criteria for assessing the harmfulness of radioactive materials and waste. In order to make the indicators understandable to a wide audience, the situations are defined to respect a minimum degree of realism. Their choice also aims to cover the main exposure routes and a diversity of contexts.

Four situations are considered, the first two of which involve the presence of an individual in a room containing a package of radioactive waste or radioactive material, whether intact or damaged. The last two situations concern the dispersion of the package in the environment and the impact on an entire local human population or the impact on an aquatic ecosystem.

The report also provides an example of the application of the method for three families of waste (vitrified HA, bituminous MAVL and FAVL ^{14}C). The annual impacts after 100 or 1,000 years are provided and proposals are made for broader deployment, making it possible in the long-term to have an indication of the harmfulness of each of the families defined in the national inventory of radioactive materials and waste [IRS 18b].

1.3.4. American classification

The American classification of radioactive waste has three classes (A, B and C) based on the maximum activity of a given radionuclide (Table 1.2).

Radionuclide	Class A	Class B	Class C
^3H	40	MC	MC
^{14}C	0.8	–	8
^{60}Co	700	MC	MC
^{90}Sr	0.04	150	7,000
^{99}Tc	0.3	–	3
^{129}I	0.008	–	0.08
^{137}Cs	1	44	4,600
All radionuclides with half-life <5 years	700	MC	MC
α emitters with a half-life >5 years	10		100
^{241}Pu	350		3,500
^{242}Cm	2,000		20,000

Table 1.2. Excerpt from the US NRC classification of radioactive waste based on maximum concentrations of radionuclides and expressed in Ci.m^{-3} (source: [BLA 01]). MC: maximum concentration (no limit for this class)

1.3.5. British classification

The British classification of radioactive waste adopts the IAEA classification into five categories by defining its own criteria for activity levels (Table 1.3).

Waste classes	Characteristics of this class
VLLW, small volume	Waste of 0.1 m ³ that can be disposed of with regular garbage if it contains less than 400 kBq of activity, as well as hospital and university waste. For waste containing carbon-14 and tritium, the activity limit is 4,000 kBq
VLLW, large volume	Radioactive waste with an upper limit of 4 MBq per ton (not including tritium) is disposed of in specified landfills. For waste containing tritium, the upper limit is 40 MBq per ton
LLW	Containing radioactive material other than that suitable for disposal with ordinary waste, but not exceeding 4 GBq per ton of waste or 12 GBq per ton of β and γ activity
ILW	Waste with radioactivity levels above the upper limits for LLW, but which does not generate heat
HLW	Wastes in which the temperature can increase significantly due to their radioactivity, so this factor must be taken into account in the design of storage or disposal facilities

Table 1.3. *The British nuclear waste classification system (source: [OJO 14, RAH 15])*

1.3.6. Russian classification

The Russian classification of radioactive waste is based on a division into three classes according to the specific activity of various categories of radionuclides (Table 1.4). The limits of the categories are high.

Category	Specific activity (Bq.g ⁻¹)			
	Tritium	Beta (except ³ H)	Alpha (except transuranium elements)	Transuranium elements
Low activity	10 ⁶ –10 ⁷	<10 ³	<10 ²	<10
Average activity	10 ⁷ –10 ¹¹	10 ³ –10 ⁷	10 ² –10 ⁶	10–10 ⁵
High activity	>10 ¹¹	>10 ⁷	>10 ⁶	>10 ⁵

Table 1.4. *Practical classification of radioactive waste in Russia (source: [OJO 14])*

1.3.7. Comparisons of the various classifications

Various comparisons can be made between the classifications of radioactive waste used by different countries.

1.3.7.1. American classification and IAEA recommendation

The classification recommended by the IAEA and that applied by the United States have no overlap (Table 1.5).

NRC	Class A		Class B	Class C	Excess C or GTCC
IAEA	VLLW	LLW	ILW		HLW

Table 1.5. Comparison of IAEA ([IAE 09a], GSG-1) and NRC ([NRC 15]) classifications (source: [NEA 16a])

1.3.7.2. Comparison between the Belgian, French and Canadian radioactive waste classifications

In Belgium, class A waste has a specific destination and class B and C waste are managed together. In France, the VLLW and LLW-SL categories are managed together, the AA-LL and HALL categories are managed together, while the FA-VL category is managed independently. For the three states, a distinction is made between current waste and historical waste [PAR 18].

	Belgium	France	Canada
Number of categories	3	5	4
Classification by lifespan and activity level	A (LLW) B (ILW) C (HLW)	TFA (VSLW) FMA-VC (LLW) FA-VL (VLLW) MA-VL (ILW) HA-VL (HLW)	LLW (LLW) ILW (ILW) HLW (HLW + spent fuel) Mining waste
Other more vague categories	NORM, T-NORM Radifer Waste from future sanitation Spent fuel Spent MOX fuel	Waste without a channel Fuel and MOX	

Table 1.6. Comparison of radioactive waste classifications in Belgium, France and Canada (source: [PAR 18]). In brackets, the equivalences with the IAEA classification from 2009 [IAE 09a]

1.3.8. Classification of sealed sources

For sealed sources, the IAEA [IAE 09a] recommends the classifications reported in Table 1.7.

Type	Half-life	Activity	Volume	Examples
VSLW	<100 days	100 MBq	Small	^{90}Y , ^{198}Au (brachytherapy)
VSLW	<100 days	5 TBq	Small	^{192}Ir (brachytherapy)
LLW	<15 years	<10 MBq	Small	^3H , ^{60}Co , ^{85}Kr
ILW	<15 years	<100 TBq	Small	^{60}Co (irradiators)
LLW	<30 years	<1 MBq	Small	^{137}Cs (brachytherapy)
ILW	<30 years	<1 PBq	Small	^{90}Sr (thickness gauges, thermoelectric generators), ^{137}Cs (irradiators)
ILW	>30 years	<40 MBq	Small but with a large number of sources	Pu, Am, Ra (static eliminators)
ILW	>30 years	<10 GBq		^{226}Ra , ^{241}Am (gauges)

Table 1.7. Examples of the use of the IAEA classification for disused sealed radioactive sources (source: [IAE 09a])

1.4. Origins of nuclear waste

Radioactive waste has multiple origins, which can be subdivided into three main sources: waste from the fuel cycle contributing to nuclear electricity (NFC, Nuclear Fuel Cycle), waste from other very varied origins (medicine, research, etc.) and waste resulting from a nuclear accident. Fuel cycle waste differs according to whether it comes from upstream or downstream plants or from nuclear power reactors in operation (Figure 1.2).

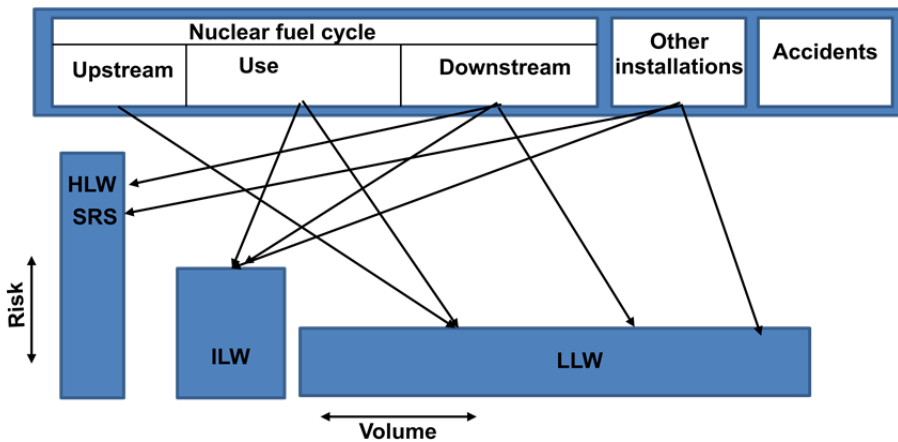


Figure 1.2. Diagram of the origins of radioactive waste (source: [OJO 14]). HLW: high-level waste; ILW: intermediate-level waste; LLW: low-level waste; NFC: nuclear fuel cycle; SRS: sealed radioactive sources. For a color version of this figure, see www.iste.co.uk/amiard/radioactive.zip

1.4.1. The main radionuclides in radioactive waste

The principal radionuclides in radioactive waste are very varied and can be classified into four categories. These are fission products (H, Se, Br, Kr, Rb, Sr, Y, Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te, I, Xe, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb and Dy), activation products (C, Cr, Mn, Fe, Co, and Ni) and heavy nuclei (U, Nb and Zr), those that are both fission and activation products (Zr and Nb), heavy nuclei (U, Np, Pu, Am and Cm) and some elements with long-lived radioactive isotopes (C, Zr, Tc, Pd, Sn, I, Cs and Sm) to which are added the five heavy nuclei elements.

1.4.2. Wastes related to the nuclear fuel cycle

A distinction should be made between two fuel cycles, the so-called open NFC and the closed NFC, the latter reprocessing spent nuclear fuel in order to reuse the extracted by-products (uranium and plutonium) in other reactors, whereas in the case of the open NFC, the spent fuel is considered as radioactive waste and therefore disposed of. A representation of the two types of fuel cycle is shown in Figure 1.3.

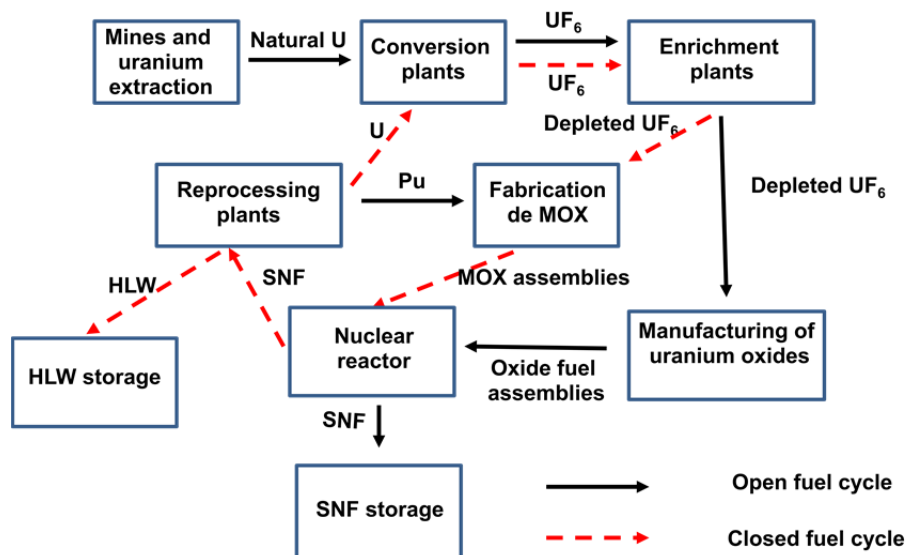


Figure 1.3. The various stages of the nuclear fuel cycles in open and closed versions (source: [OJO 14]). HLW: high-level waste; MOX: mixed oxide; NFC: nuclear fuel cycle; Pu: plutonium; SNF: spent nuclear fuel; U: uranium; UF₆: uranium hexafluoride. For a color version of this figure, see www.iste.co.uk/amiard/radioactive.zip

The number of states reprocessing civilian spent fuel in 2013 was still six (China, France, India, Japan, the United Kingdom and Russia) with a theoretical annual reprocessing capacity of 5,900 tons to be increased to 6,700 tons [OJO 14]. In 2020, the United Kingdom gave up reprocessing and Japan has had its plants shut down for many years.

The chemical and radioactive composition of HLW varies greatly from state to state. Thus, for transuranium elements, the quantities present in HLW, expressed in g.L⁻¹, are 2.0 for the British Magnox reactors, 5.1 for the waste from the La Hague reprocessing plant in France, 7.6 for the WIP (Waste Immobilization Plant) in India, 12.6 for the waste from the Tokai reprocessing plant in Japan and <0.1 for American Hanford waste. Similarly for fission products, the quantities expressed in g.L⁻¹ are 87.0 at La Hague, 1.1 at the Indian WIP, 49.0 for the Japanese Tokai plant and <2.5 for the Hanford waste. This can be explained by the characteristics of the reactors and nuclear fuels used, as well as by the cooling methods used and the reprocessing technologies [OJO 14].