Human-made radioactivity

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Note

In this document the references are coded by Q-numbers (e.g. Q6). Each reference has a unique number in this coding system, which is consistently used throughout all publications by the author. In the list at the back of the document the references are sorted by Q-number. The resulting sequence is not necessarily the same order in which the references appear in the text.

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Summary

Chapter 2 describes briefly the nuclear process chain from cradle to grave: from mining uranium ore through final disposal of the last radioactive waste generated by a particular nuclear power plant.
As it turns out, the back end of the process chain is not finished: the crucial components of the nuclear tail still do not exist. This situation is comparable with a household in which dinner has been cooked and enjoyed, but washing the dishes and cleanup of the house is left for future residents of the house. Finishing the nuclear chain in a sound and safe way is not a matter of advanced technology, but a matter of economic effort.

Unique feature of nuclear power is the generation of human-made radioactivity. The amount of radioactivity leaving a reactor is about a billion times the quantity entering the reactor. A 1 GWe nuclear power plant generates each year as much radioactivity as 1000 exploded nuclear weapons (Hiroshima bomb). The world inventory of human-made radioactivity is more than 10 million nuclear bomb equivalents. By the fission process in a nuclear reactor, dozens of different kinds of radionuclides are generated, each kind having its own physical and chemical properties and each having its specific decay mode and half-life.
The risks of dispersion of human-made radioactivity into the human environment increase exponentially with time for a number of reasons, one of them being the consequences of the Second Law of thermodynamics.

The radioactivity generated in a nuclear power plant ends up in different waste streams. The bulk of the radioactivity is contained in spent fuel. Due to the high level of radioactivity in a small volume, spent fuel is heat-generating and has to be cooled during at least ten years, to avoid melting. The remainder of the radioactivity is distributed over large volumes of materials: not only mining waste and so-called operational waste, but also dismantling waste, a systematically forgotten highly hazardous radioactive waste stream of the nuclear energy system.
A substantial part of the human-made radioactivity is released into the biosphere, partly as authorized discharges, partly as a result of leaks and accidents.

The nuclear industry distinguishes several categories of radioactive waste, according to the intensity of radiation (in practice always γ-radiation) measured outside a waste container. This classification has a strong economic component and is often different from country to country. Due to the decay of radionuclides, the radioactivity of nuclear waste decreases over time. Spent fuel remains highly hazardous for thousands to millions of years. Other radioactive wastes always contain long-lived radionuclides and will remain hazardous also during that timescale, albeit at a much lower activity level. Actinides are considered to be the most hazardous radionuclides in spent fuel, but their presence in other, less hazardous qualified wastes, cannot be excluded.

Radioactivity cannot be destroyed, nor made harmless to man. Consequently the only way to minimize the hazards posed by the human-made radioactivity is to isolate it from the human environment and from the biosphere as long as possible. The best way to achieve a durable isolation from the biosphere is to store all radioactive waste in repositories deep in geologically stable formations. Two kinds of repositories are envisioned: one for heat-generating waste and the other for other radioactive waste. The former comprises a system of tunnels and galleries with separate holes, each containing one canister with spent fuel, the latter consists of tunnels and large cavernes, to be filled with containers of operational and dismantling waste.
The generic concept of a geologic repository is now more than 30 years old, but at the time of writing (2019) no such repository exist in the world. In fact, all human-made radioactivity is still present in mobile state in the biosphere. To dispose of the existing global backlog of spent fuel at least six very large geological repositories are needed, and from then on every four years a new one has to become operational.

Spent nuclear fuel consists chiefly of unfissioned uranium and contains some 3.5% fission products and some 1,5% actinides which are formed from uranium by neutron capture. Both the fission products and the actinides, among which plutonium, are highly radioactive. In addition to direct disposal in a geologic repository, the nuclear industry proposes two other options to manage the spent fuel: reprocessing + vitrification and partitioning (an advanced version of reprocessing) + transmutation.
Reprocessing of civil spent fuel has been developed to extract plutonium and unfissioned uranium, for recycling in closed-cycle (breeder) reactors. As the concept of a closed-cycle reactor proved to be technically unfeasible, reprocessing, an extremely polluting and expensive technology, became essentially superfluous.
Reprocessing + vitrification as a solution to the spent fuel issue is a fallacy: instead of volume reduction of the highly radioactive waste, the human-made radioactivity is distributed over large volumes of materials and moreover an appreciable part of the content of spent fuel is discharged into the biosphere. Moreover reprocessing creates high risks of proliferation and terroristc use of plutonium and neptunium-237.
The concept of partitioning + transmutation, as a solution to the spent fuel issue and as a way to generate more useful energy form natural uranium than the current generation reactors, is based on several fallacies. The most important flaw in the concept is the ignorance of the consequences of the Second Law of thermodynamics.

A view on radioactive waste quite different from this report is communicated by the nuclear industry to the policy makers and the public. Firstly the nuclear industry denies nuclear waste being a problem, despite the fact that all radioactive waste ever generated is still present in vulnerable temporary storage facilities, essentially mobile in the human environment.
Secondly the nuclear industry asserts:

> "Nuclear power is the only large-scale energy-producing technology which takes full responsibility for all its wastes and fully costs this into the product."

This statement is little short of a lie, because in all countries, as far as known, the management of spent fuel and dismantling waste are the responsibility of the governments and become a public charge, tens of years after closedown of a particular nuclear power plant and possibly even after discontinuance of the owning firm.

This study concludes that all radioactive wastes, including dismantling waste, should be permanently stored in a deep geologic repository.

This report addresses the generation of human-made radioactivity. Other aspects of the nuclear waste problem are addressed in separate reports retrievable on this website.

1 Radioactivity, some basics

Nuclear reactions

To understand some unique features of nuclear process chain, connected with the radioactivity generation, a short description of the nuclear reactions taking place in an operating reactor may be helpful.

Fuel elements of a LWR consist of bundles of thin tubes of Zircalloy (an alloy of zirconium and a few percent other metals, e.g. tin), containing pellets of uraniumoxide UO2. Actually, the term 'fuel' is wrong, because the uranium in the reactor does not burn with oxygen, like coal in air, but its atoms fission. Yet, fissioning of uranium often is called 'burning' of uranium and fissionable material is called 'fuel'.

Nuclear reactions are processes in which the nuclei of atoms change and are completely different from chemical reactions, in which the changes are confined to the electron clouds of atoms and the nuclei stay unchanged throughout. Three kinds of nuclear reactions are taking place in an operating reactor:

- fission of heavy atoms
- neutron capture (activation)
- radioactive decay.

During the fission process in the reactor, an intense neutron flux exists in the reactor. A part of the free neutrons maintain the fission process, the nuclear chain reaction, another part is captured by uranium atoms without fission and the remaining neutrons escape from the core and are captured by the materials the reactor and the surrounding biological shield are made of.

Fission

When a uranium nucleus fissions, two nuclei of lighter atoms, the fission products, plus two or three free neutrons come into being. The fission products represent a large number of different elements: a major part of the Periodic System is represented in spent fuel. A part of the new atoms is highly radioactive. As a matter of fact, the radioactivity of the fuel elements soars with a factor of one billion by the generation of radioactive fission products.

Activation

The process of neutron capture is called activation, because most non-radioactive atoms which catch one or more neutrons become radioactive. Two activation processes are important: neutron capture by uranium atoms and neutron capture by atoms of the coolant and construction materials.

By neutron capture and following decay processes, a part of the uranium-238 atoms is converted into plutonium-239. This is a desired proces, because plutonium-239 nuclei are fissile in a LWR and uranium-238 not. By subsequent neutron capture, plutonium nuclei are converted into nuclei of americium, curium and higher trans-uranium elements, an unwanted process. That group of elements is called actinides or trans-uranium (TRU) elements. All are strongly radioactive and most of them alpha-emitters. Sometimes the trans-plutonium nuclides are referred to as 'minor actinides', a term which conceals the serious hazards they pose and only denotes the minor contribution in the total mass of spent fuel. In effect, the actinides often are considered a major problem of the radioactive wastes.

By activation reactions the coolant and the construction materials of the fuel elements, reactor and surrounding structures become strongly radioactive, inevitable.

Isotopes

Atoms are composed of a nucleus with a positive electric charge surrounded by electrons with a negative charge. The negative charge equals the positive, so the atom is electrically neutral. The nucleus consists of protons (positive charge) and neutrons (neutral). The number of protons determines to which chemical element the atom belongs. The chemical properties of an atom are determined by the number of protons. The number of neutrons may vary; atoms with an equal number of protons but a different number of neutrons in the nucleus are called isotopes. The chemical properties of isotopes are identical. The nuclear-physical properties of an atom are partly determined by the number of neutrons. Some isotopes have an unstable nucleus.

Radioactive decay

Radioactivity is the phenomenon that unstable nuclei of atoms (radionuclides) spontaneously decays into another kind of atom, coupled with the emission of nuclear radiation: alpha, beta and/or gamma radiation. Alpha radiation consist of alpha (α) particles: helium-4 nuclei (2 protons + 2 neutrons) which are ejected from the decaying nucleus at very high speed. Beta radiation consist of beta (β) particles: electrons ejected from the nucleus at very high speed. Gamma radiation consists of gamma (γ) rays, very energetic electromagnetic rays and much more penetrating than X-rays.
The decay product, also called the decay daughter, can be radioactive in itself or can be stable. In nature on earth a few radioactive kinds of atoms occur in low concentrations. Important with respect to nuclear power are the elements uranium and thorium, which are formed billons of years ago in supernova explosions. These radionuclides decay via a series of other radionuclides into stable lead or bismuth atoms. During fission of uranium large amounts of radioactive isotopes of nearly all known elements are formed.

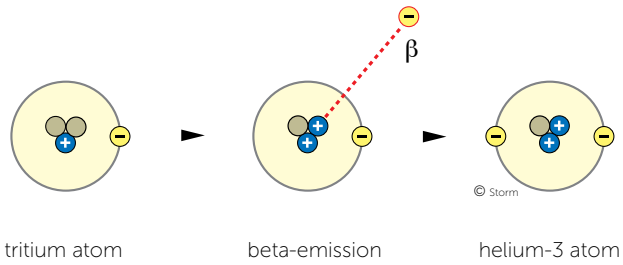

Figure 1.
Radioactive decay of tritium. Tritium, symbols T, ^{3}H or H-3, is a heavy isotope of hydrogen, with one proton and two neutrons in the nucleus. When a tritium atom decays, it emits a beta particle (an electron) at high speed. After decay the nucleus contains two protons and one neutron, the nucleus of a helium-3 atom, which captures a second electron and becomes a neutral helium-3 atom. The sums of electric charges remain constant and a minute fraction of the mass is converted into energy.

Ionising radiation

Nuclear radiation is often called ionising radiation, because it strongly interacts with matter forming ions. Ionising radiation is harmful to living organisms, for it destroys biomolecules. Alpha and beta radiation can be blocked by thick paper respectively aluminum foil, so these rays may seem not very harmful to man. However radionuclides radiating alpha or beta rays inside the human body are extremely dangerous, because the living cells are not protected by the skin or clothes. A dose of only a few nanograms of the alpha-emitter polonium-210 in the human body is lethal.
A complicating factor is that alpha and beta radiation are not detectable by hand-held counters, which can

only detect gamma rays. Radionuclides that emit weak or no gamma rays are invisible to these detectors. A number of biologically very active radionuclides fall within this category, such as tritium (radioactive hydrogen) and carbon-14 (radioactive carbon).

Figure 2.
Symbol of nuclear radiation. This pictogram symbolizes three kinds of lethal nuclear radiation: alpha (α), beta (β) and gamma (γ) radiation.

Half-life

The rate of radioactive decay is characteristic to each kind of radionuclide and cannot be decelerated or accelerated by any means. Radioactivity cannot be destroyed nor made harmless to man and other living organisms. Radionuclides occurring in nature, such as uranium and thorium, have very long half-lifes measured in billions of years. These nuclides have been formed in stellar explosions long before the Earth came into being. Human-made radionuclides have much shorter half-lifes, ranging from seconds to millions of years. The specific radioactivity of a radionuclide (measured in becquerel per gram, Bq/g) is higher as the half-life is shorter.

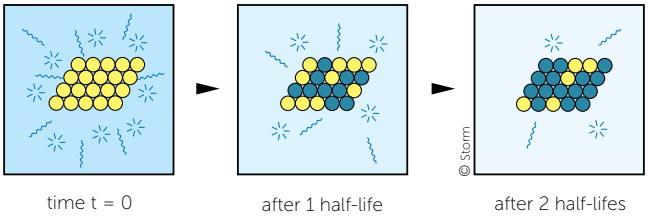

Figure 3.
Decay of a radionuclide. One half-life period after creating of a given amount of a certain radionuclide at time $t = 0$, half of the radionuclides has decayed into another kind of nuclide, called the daughter nuclide. In most cases the decay doaughter is a non-radioactive, stable nuclides. During the next half-life period half of the remaining radionuclides decay, and so on. The total mass of matter remains almost constant during the decay process. The daughters of a number of heavy radionuclides, e.g. uranium and plutonium, are radioactive in itself.

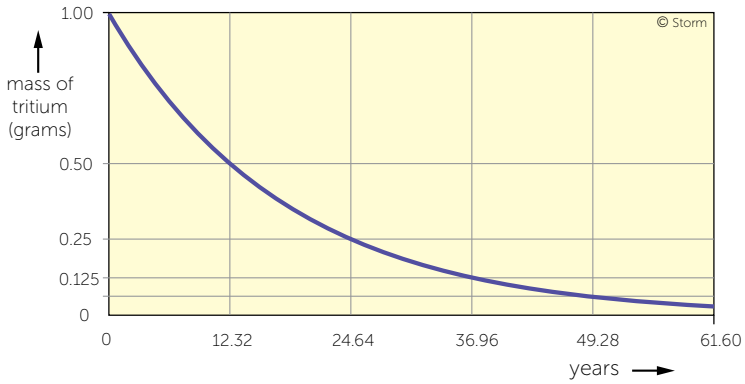

Figure 4.
The mass of a given amount of tritium as function of the time. After one half-life half of the initial number of tritium atoms has decayed into helium-3 atoms. During the second half-life period half of the remaining tritium atoms decay. Radioactive decay is a stochastic process.

Nuclear bomb equivalents

Nuclear power generates immense amounts of radioactivity, irrevocably and irreversibly. During fission of uranium atoms many dozens of different kinds of radioactive atoms are coming into being, called the fission products. In addition non-radioactive construction materials become radioactive by neutron radiation. The amount of human-made radioactivity is a billion times the radioactivity of the fresh uranium entering the reactor.

One nuclear reactor generates each year an amount of radioactivity equivalent to roughly 1000 nuclear bombs of the yield of the Hiroshima bomb. All radioactive wastes ever generated during the nuclear era are still stored in temporary storage facilities. These facilities are leaking all kinds of radioactivity into the environment at an increasing rate, due to the unavoidable degradation of the materials and structures of the containing facilities, and are vulnerable to natural disasters, accidents and terroristic attacks.
Once generated, radioactivity cannot be influenced by any means. The radioactivity resulting from nuclear power decreases by natural decay only. For some human-made radionuclides the decay rate can be measured in seconds to hours, for other time scales of years to millions of years are involved.

2 Unique feature of nuclear power

Nuclear power, fission as well as fusion, is in one respect distinct from all other energy systems, namely, the generation of radioactivity. A nuclear reactor is a generator of heat and radioactivity, simultaneously, inextricably and irreversibly. This unique feature has far-reaching consequences.
The flow of radioactivity in the nuclear process chain starts with the mobilization of natural radioactivity of the uranium ore and multiplies a billionfold by the generation of man-made radioactivity in the reactor. Unavoidably a part of the mobilized and man-made radioactivity will be released into the environment. Purpose of the back end of the nuclear chain is minimalize the discharges of radioactivity into the human environment for unprecedented long times.

Radioactivity cannot be destroyed, nor be made harmless to man, so the sole means to limit the harm inflicted to the population by nuclear power is to immobilize the generated and mobilized radioactivity as soon as possible and as effective as possible. A safe completion of the nuclear process chain exists only in cyberspace.
This report addresses the generation of human-made radioactivity, how it should be handled and how it actually being handled.

3 Nuclear process chain, an unfinished system

A nuclear power plant is not a stand-alone system, but is part of a chain of industrial processes, each of which is vital for the generation of electricity from uranium ore. The whole complex is coined the 'nuclear process chain' or the 'nuclear energy system' and comprises three main sections: the front end processes, the mid-section and the back end processes.

- *Front end processes*

Nuclear fuel is not found ready to use in nature, like coal or oil, but has to be produced from uranium ore, by means of a number of industrial processes. Jointly these processes – from ore to fuel – form the front end or head of the chain and comprise: mining + milling (extraction of uranium from its ore), conversion of uraniumoxide from the mine into uranium hexfluoride, enrichment and fuel fabrication.

- *Mid-section*

The mid-section of the nuclear chain encompass the construction of the nuclear power plant plus the operation and maintenance of the plant during its operational lifetime. Maintenance includes refurbishment of the plant. At the end of its operational lifetime nearly all components of a nuclear power plant have been replaced at least once, except the reactor vessel and the reactor building.

- *Back end processes*

The back end processes, also called the tail of the nuclear process chain, comprise all processes needed to effectively isolate the radioactive waste generated by the nuclear energy system from the human environment. The back end processes include cleanup, decommissioning and dismantling of the radioactive part of the nuclear power plant after closedown, and of other radioactive facilities of the nuclear chain.

Nuclear chain as it ought to be

A simplified outline of the nuclear chain from cradle to grave is illustrated by Figure 5. A full description of

the complete nuclear process chain can be found in Parts B and C of [Storm & Smith 2008] Q6. The nuclear process chain starts with the extraction of uranium from the Earth's crusts and ends with the final disposal of the human-generated radioactivity into the biosphere. Disposal of the radioactive wastes in a deep geologic repository is generally considered to be the least risky way of dealing with the anthropogenic radioactivity. Report m32 *Geologic repositories* addresses the concept of the deep geologic repository in more detail.

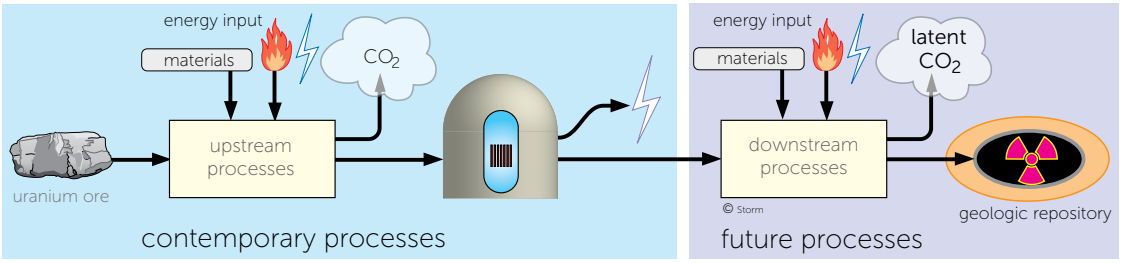

Figure 5
Simplified outline of the nuclear process chain, as it ought to be from cradle to grave

Like any industrial process chain, the nuclear process chain may be compared with a common, daily chain of housekeeping activities: collecting the ingredients + cooking the meal, enjoying the meal and washing the dishes + clearing the mess.

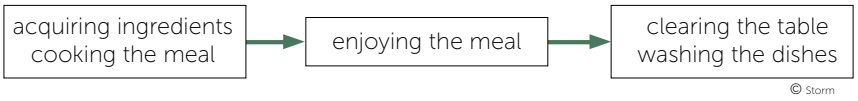

Figure 6
A metaphoric representation of all industrial production processes, including the generation of electricity by nuclear reactors

The back end processes turn out to be be ill-defined, the most important processes are even non-existent. Most noticeable is the global absence of the construction and operation of geologic repositories. Because of the huge amounts of radioactivity involved, the back end processes pose the majority of the health risks originating from civilian nuclear power. Or, as the old Romans said: *in cauda venenum*.

Nuclear chain: the current practice

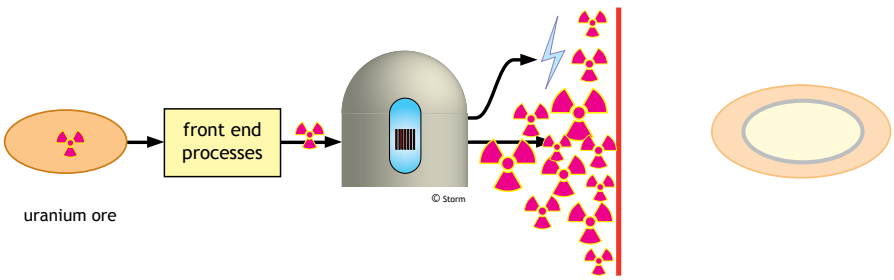

Figure 7
The practice: the radioactive waste from more than 60 years of nuclear power is piling up in temporary storage facilities and is still mobile in the human environment. The barrier preventing the human-generated radioactivity from adequate isolation from the human environment is not a technical one, but a paradigmatic one.

As common in daily life, nobody likes to do the dishes and clean up the mess. The aftermath of the nuclear meal turns out to be postponed indefinitely. The dirty dishes are piling up in the kitchen and dining room, our living environment. Why do governments, industry and media hide their eyes from this mess?
After more than 60 years of nuclear power the radioactive wastes ever generated are still awaiting definitive processing, while stored in temporary, above-ground storage facilities, a situation getting more unsafe day by day. Why has not a single piece of the radioactive waste of the nuclear era been effectively isolated from the biosphere?
On closer consideration this question turns out to be not primarily a technical problem, although a demanding task, but a paradigmatic and financial problem: the nuclear legacy.

4 Mobilisation and generation of radioactivity

Mobilising natural radioactivity

Uranium is a radioactive metal found in nature in various chemical forms in uranium ore. In uranium ore strata, uranium and its many radioactive decay daughters are bound in chemically stable minerals. This is not to say uranium-bearing rock is harmless to man, not at all.
When uranium ores are disturbed to extract the uranium, the element is brought out of its geologic confinement into the environment and is chemically mobilized. The uranium isotopes are chemically separated from their their decay daughters and converted into nuclear fuel. In addition, the separated radioactive daughters of uranium are dumped as uranium mill tailings in huge ponds and spoil heaps. From then on the radioactivity from the uranium ore is mobile.

Generating radioactivity

Human-made radionuclides come into being during the fission process in the reactor. Three different groups of the artificial radionuclides are commonly distinguished, according to their origin:

- Fission products: the light atoms originating from the fission of the heavy uranium and plutonium atoms. Atoms of nearly all chemical elements are present in the mix and a part of the fission products are highly radioactive.
- Transuranic actinides: atoms heavier than uranium, which are formed from uranium atoms by neutron capture. These elements, for example plutonium and americium, do not occur in nature and are highly radioactive and highly toxic.
- Activation products. All non-radioactive materials exposed to neutron radiation from the fission process become radioactive by neutron capture; examples are the nuclear fuel cladding and the reactor vessel itself.

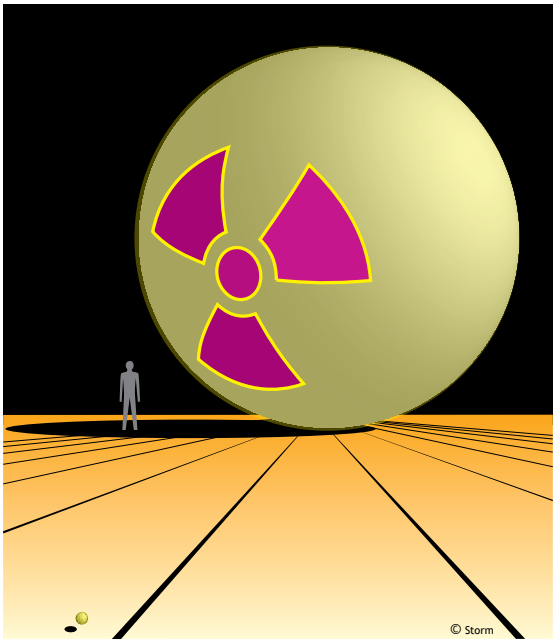

Figure 8
As a result of the fission process the radioactivity of the nuclear fuel irreversibly increases a billionfold. The pea in the foreground (diameter 1 cm) represents the original radioactivity of the nuclear fuel, the sphere in the distance with a diameter of 10 meter represents the radioactivity of the same amount of fuel 150 days after shutdown of the reactor. Based on data from: [Bell 1973] Q264 , [Hollocher 1975] Q262 and [JPL-77-69 1977] Q263.

During the fission process the radioactivity of the nuclear fuel increases a factor *billion*. The generation of these immense quantities of radioactivity is irreversible. Radioactivity cannot be destroyed nor can be made harmless to man. Radioactivity only decreases by natural decay, a process which cannot be controlled by man. The radiation emitted by radioactive substances is hazardous to man, especially when the radionuclides enter the body, via food, water or inhalation. This issue will be addressed in more detail in reports m11 *Health effects of radioactivity* and m17 *Pathways of radioactive contamination*.

The highly dangerous spent nuclear fuel is unloaded from the reactor and transported to temporary storage facilities, usually on the site of the nuclear power plant. In any case the radioactivity leaving the reactor is physically mobile and mobile radioactivity poses a direct threat to human health.

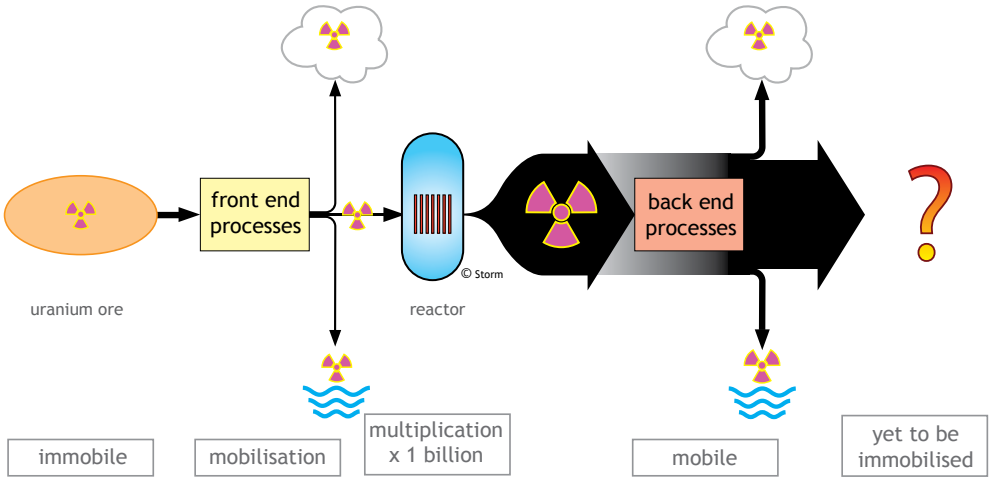

Figure 9
Symbolic presentation of the radioactivity flow through the nuclear system. The flow starts with the mobilisation of natural radioactivity and multiplies a billionfold by generation of radioactivity in the reactor. Unavoidably a part of the mobilised radioactivity will be released into the environment. A safe immobilized end of the nuclear chain still exists only in cyberspace. What quantities of the radioactive materials leaving the reactor will end up in the environment?

Nuclear bomb equivalents

Each nuclear reactor of 1 GWe power produces each year an amount of radioactivity roughly equivalent with 1000 exploded nuclear bombs of 15 kilotonnes, about the yield of the Hiroshima bomb. A nuclear reactor generates relatively more long-lived dangerous alpha-emitting radionuclides (actinides) than an exploding nuclear bomb, so the radioactivity from a reactor is more dangerous than an equivalent amount from a nuclear explosion.

Once generated, radioactivity cannot be influenced by any means. The radioactivity resulting from nuclear power decreases by natural decay only. For some components of the man-made radioactivity the decay rate can be measured in seconds to hours, for other components timeframes of years to millions of years are involved.

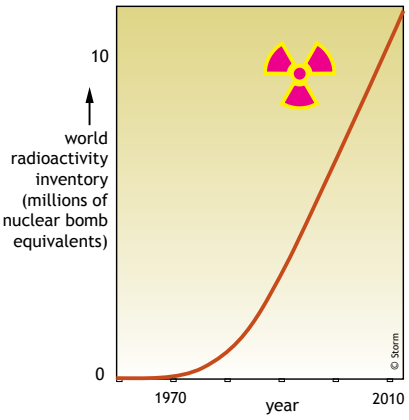

Figure 10

All man-made radioactivity still exists in a mobile form in the human environment. The world inventory of man-made radioactivity passed the 10 million nuclear bomb equivalent mark in 2010 and is rising nearly linearly with 370 000 nuclear bomb equivalents a year.

Exponential growth of dispersed human-made radioactivity

All radioactive wastes ever generated during the nuclear era are still stored in temporary storage facilities. These facilities are leaking all kinds of radioactivity into the environment at an increasing rate, due to the unavoidable deterioration of the materials and structures of the containing facilities, and are vulnerable to natural disasters, accidents and terroristic attacks.

Ever since the first nuclear reactors started operation in the 1940s, the definitive solution to the radioactive waste problem has been postponed to the future. Spills from corroding storage tanks and waste containers are polluting watersheds, rivers and sea. A systematically underrated aspect of civilian nuclear power is the enormous size of the radioactive waste problem and its exponential growth over time.

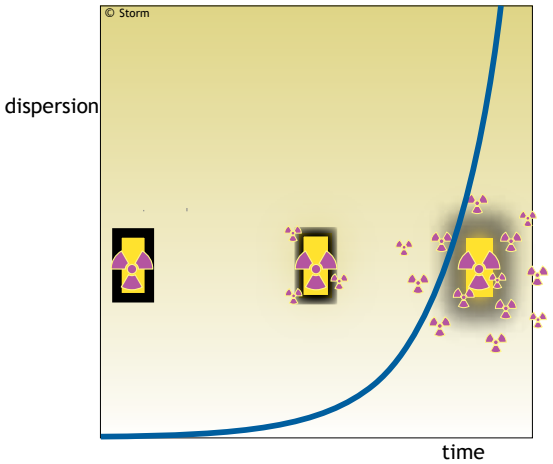

Figure 11

Increasing dispersion of radioactive materials into the environment over time, due to deteriorating containments of the radioactive materials as a consequence of the Second Law. The dispersion rate will increase exponentially, because the corrosion of the containments will progress exponentially. Self evidently the exponential dispersion rate causes an exponential increase of health hazards over time, even if the amount of radioactivity would remain constant.

The exponential growth of the radioactivity problem has several causes:

- Each year an amount of radioactivity equivalent to some 370 000 nuclear bomb explosions is added to the world radioactivity inventory, embodied in the spent fuel from civilian nuclear reactors. Since the 1970s the world inventory has increased with a factor 1000 and the proposed solutions still exist only on paper or in cyberspace.
- The dispersion rate of the anthropogenic radioactivity increases progressively over time caused by the inevitable ageing and deterioration of the materials and constructions of the containments, phenomena governed by the Second Law of thermodynamics.
- During an economic recession society has a declining ability to cope with nuclear health risks. The safe storage of radioactive waste requires vast amounts of energy, materials and economic effort. Conflicts may surface between short-term economic priorities and and long-term, less visible health effects. We return to the economic aspects in reports m05 *Downplaying and denial of health effects* and m34 *Conflict of interests, flexibility of regulations*.

Mobile radioactivity and the Second Law

Time is a formidable ennemy of the safety of nuclear power. The longer the definitive isolation and immobilisation of radioactive waste in a safe geologic repository is postponed, the more likely radioactivity will be spread into the environment and the more efforts will be necessary to prevent severe nuclear accidents. The risks will increase with time due to the progressive and irrevocable loss of quality of and reliability of materials and structures by a number of spontaneous mechanisms. The degrading character of spontaneous processes follows from the Second Law of thermodynamics. See also the notion of engineered safety, to be addressed in reports m21 *Nuclear safety* and m38 *Nuclear power and the Second Law*.

5 Radioactive waste

Classification

The radioactive waste streams of the nuclear system can be grouped in several ways. Important with regard to the final disposal is the distinction between radioactive waste that generates heat and waste that does not. Heat-generating waste has a very high specific radioactivity. The heat, produced by the decay of the radionuclides, may be sufficient to melt the waste, if not cooled adequately. This property may cause serious health risks and creates problems for permanent disposal of this kind of waste.

Spent fuel generates heat for long times after removal from the reactor. If spent fuel is reprocessed, the radioactive contents are redistributed over other heat-generating waste volumes: solids (cladding hulls and insolubles), liquids and vitrified waste, containing the bulk of the actinides and a major part of the fission products. For reason of its high specific radioactivity spent fuel or fractions of it are usually referred to as High Level Waste (HLW) or High Level Long-lived Waste (HLLW), for reason of the long half-lifes of the radionuclides in the waste.

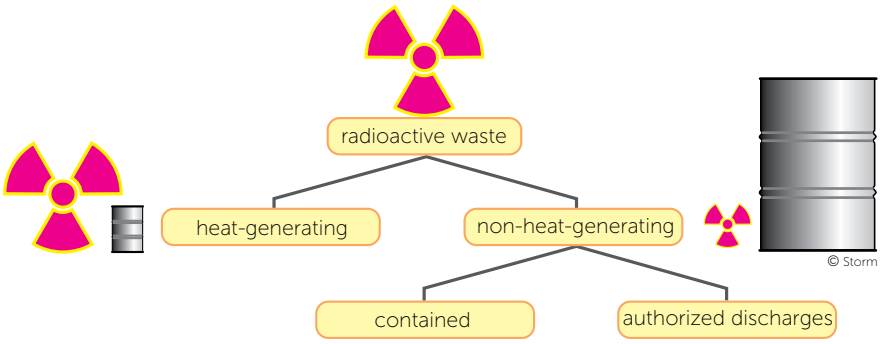

Figure 12

Schematic classification of the radioactive wastes streams produced by the nuclear energy system. Spent fuel has a relatively small volume but a very large radioactivity inventory, the other waste streams have large volumes and a smaller radioactivity inventory than spent fuel. A part of these other waste is routinely discharged into the environment.

Other radioactive waste streams from the nuclear chain generate no significant heat due to a much lower specific radioactivity than spent fuel. These waste streams comprise:

- mining waste,
- operational waste from all industrial processes of the nuclear chain, including operation and maintenance of the reactor (except spent fuel),
- dismantling waste.

A part of these waste streams is routinely discharged into the human environment (gaseous and liquid effluents), the other part is packed into containers to be stored somewhere. The released radioactive materials include the mining waste, the light water coolant and effluents from all processes of the nuclear chain, partially due to unavoidable technical imperfections, partially for economic reasons (more see report m34 *Conflict of interests, flexibility of regulations*.

In addition there are unauthorized discharges frequently occurring, often unnoticed for long times, due to sloppy maintenance and poor inspections. Another way radioactive waste is entering the human environment is illegal trade and smuggling of valued but radioactive materials from nuclear installations. The amounts of unauthorized discharges are usually unknown, uncontrolled, but may be substantial.

The radioactive wastes other than the heat-generating wastes are classified into various categories. This classification is not internationally standardised, so different countries may use different definitions. The standards may be applied with some flexibility under economic pressure.
Based on the gamma-radiation level measured outside of a waste container the wastes are broadly classified Low-Level Waste (LLW) and Intermediate-Level waste (ILW), sometimes with subcategories. Some countries (e.g. USA) draw additionally a distinction between alpha-bearing waste and non-alpha-bearing waste, based on the presence of alpha-emitting radionuclides in the waste. Other countries (e.g. France) make a distinction between very short-lived (VSL), short-lived (SL) and long-lived (LL), based on the half-lifes of the radionuclides supposed to be present in the waste. See also the description in [WNA 2012a] Q540. Report m31 *Industrial views on radioactive waste* addresses the industrial view in more detail. Report m40 *Radioactive waste management, future CO_2 emissions* addresses the required actions to manage the nuclear legacy as safe as possible.

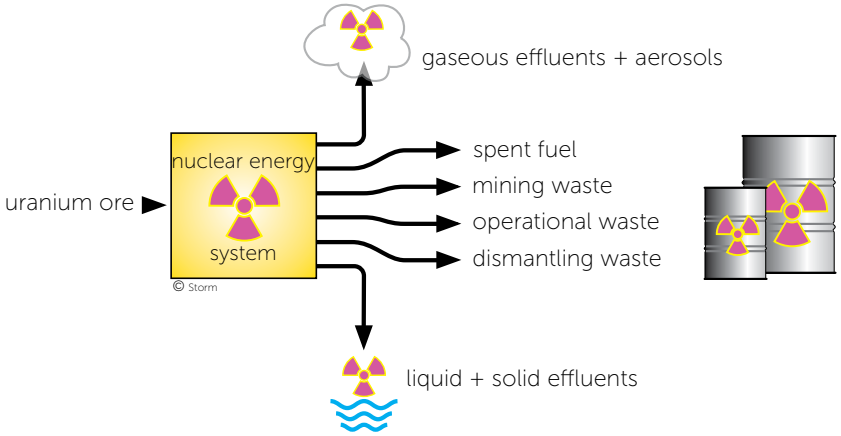

Figure 13
Outline of radioactive waste streams of the nuclear energy system, based on the origin of the waste.

Radioactive materials

A nuclear power plant is not a stand-alone system. Like most industrial production systems, the nuclear energy system from cradle to grave consists of a sequence of industrial processes comprising three sections: the front end processes, the production process itself and the back end processes.

- The front end of the nuclear chain comprises the processes to produce nuclear fuel from uranium ore: uranium recovery from the earth's crust, refining + conversion, enrichment, and fuel element fabricatrion.
- The midsection encompasses the construction of the nuclear power plant plus its operation, maintenance and refurbishments (OMR).
- The back end comprises the processes needed to manage all radioactive materials generated by the process chain, including dismantling of the radioactive parts of the power plant after final shutdown, and to minimize the hazards posed by these materials.

The full nuclear process chain from cradle to grave will be briefly discussed in report m18 *Life-cycle nuclear CO_2 emissions*.
In the reactor the radioactivity of the nuclear fuel increases a billion-fold due to the generation of fission products, plutonium and other actinides, and activation products. Significant amounts of radioactive materials escape into the environment during operation of the reactor, due to authorised routine disacharges, leakages, spills and accidents,
By far the largest part of the human-made radioactivity leaves the reactor contained in the spent fuel. In

Figure 14 this is represented by a separate pile. Most activation products are embodied in the reactor vessel + associated equipment and in the surrounding structures. The activated construction materials represent another pile of radioactive materials.

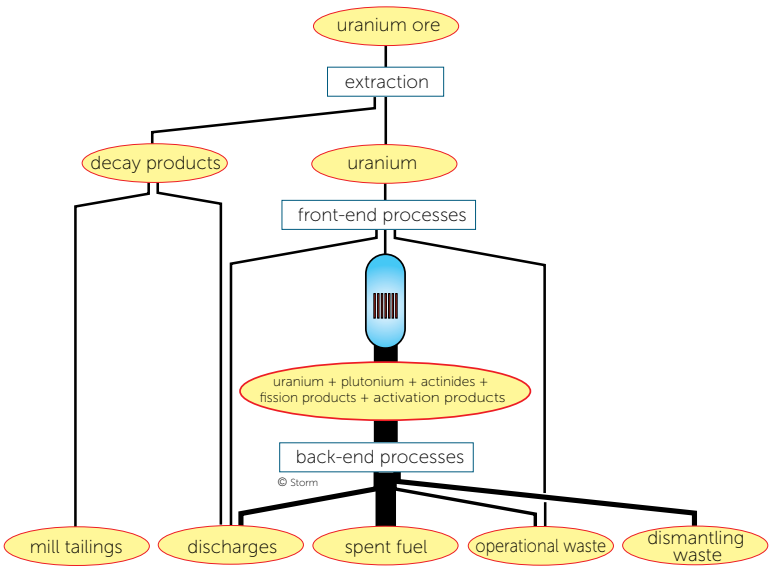

Figure 14

Outline of the generation of the radioactive nuclear legacy. The nuclear energy system generates four 'piles' of contained radioactivity, in addition to an appreciable amount of radioactivity which is discharged into the environment (air, water and soil). Details are explained in the text. Safe and definitive management of the generated radioactivity is still an unfulfilled issue, symbolised by the query.

Mobilisation of natural radioactivity

In the front end processes of the once-through nuclear chain (without plutonium recycling), from uranium ore to nuclear fuel, only naturally occurring radioactivity is involved: uranium and its decay daughters. At the mine uranium is separated from its radioactive decay products and other elements present in the ore, that end up in the waste streams of the mine (mill tailings), together with a part of the uranium, because separation processes never go to completion. All these radionuclides are in chemically mobile and reactive form. This waste stream of mobilised natural radioactivity is discharged into the biosphere. The radionuclides enter the groundwater and are also dispersed as dust and fine particulate matter, blown by the wind over vast distances. Apart from their radiological toxity uranium and its decay products are also chemically toxic. Often uranium ores contain also thorium and its radioactive decay products, that are released into the environment in the same way.

Mining waste

The reference reactor in this study, corresponding with the presently operating newest reactors, consumes some 5200 metric tons of natural uranium during its operational lifetime. Assumed that this mass is extracted from uranium ore at a grade of 0.1% uranium (the present world average is something between 0.1 and 0.05% U), then 7 million tons of ore have to be mined and processed, at an extraction yield of 80% (the current practice) to extract 5200 tons of uranium. After extraction the finely ground ore is dumped as mill tailings in large ponds, together with an estimated 0.1 million tons of chemicals used in the extraction

and some 4 million cubic meters of contaminated fresh water. These figures are based on data from the Ranger mine in Australia [ERA-AR 2005] Q321 and [ERA 2006] Q320.

Uncertainties

The classification according half-lifes rises a few basic questions. Short-lived waste, according to the French standards, contains radionucludes with half-lifes of ≤ 31 years. After ten half-life periods the radioactivity of an amount of radionuclides is reduced to 0.001 of the original amount. With a half-life of 31 years that would mean a period of 310 years.
Firstly, are three centuries a short period in the rapidly changing world of our times?
Secondly, is 0.001 of an initial amount of a dangerous radionuclide harmless?
On which empirical evidences are these assumptions based?
In our view it is practically impossible to distinguish accurately between the hazards posed by radioactive waste in a given waste container, only based on the radiation level measured outside of the container. After transport and years of temporary storage knowledge of the exact origin and content of every waste container likely will be lost.
Radioactive materials from medical sources and research laboratories generally will have a well-known radioisotopic composition. However, the composition of radioactive waste from the nucleair chain mostly will be complicated, variable and hardly retrievable, particularly the very large waste volumes resulting from:

- operation of nuclear power plants
- operation ofreprocessing plants
- cleanup of nuclear power plants after closedown, in preparation to dismantling
- cleanup of reprocessing plants and other nuclear installations, including radioactive waste storage facilities
- dismantling of the nuclear power plant and of other nuclear installations
- cleanup and dismantling of reprocessing plants.

How accurate are the measurements?
How independent are the institutions and inspectors who have to uphold the regulations?
Which warranty has the public that the way of classification and management of the radioactive wastes is not determined by financial considerations? Definitive storage of very low-level short-lived waste would be much cheaper than the definitive storage of waste containing long-lived nuclides and/or alpha emitters.
In addition the hazardous wastes are often of unknown radioisotopic composition, or contain a wide gamut of different radionuclides. Technically it is impossible to keep short-lived and long-lived radionuclides separated.
Another issue is that the physical integrity of the waste containers cannot be guaranteed. The number of leaking waste containers will inevitably rise with time, due to ageing and deterioration of the materials, one of the consequences of the Second Law of thermodynamics.

6 Radioactivity of spent nuclear fuel

Composition

Fresh nuclear fuel consists of uraniumoxide, packed in thin tubes of zircalloy. The tubes are bundled into fuel elements. The uranium has been enriched in the fissile isotope uranium-235. Fresh nuclear fuel is weakly radioactive due to the radioactivity of the uranium.
During the fission process in the reactor fissile nuclei are fissioned and the resulting fission products cumulate within the fuel pins. The fission products poisen the fuel, because they absorb the neutrons required for a sustained fission process. When the content of fission products increases above a certain level, while the content of fissile nuclei deceases simultaneously, the fission process cannot be sustained in a reliable way anymore and the fuel elements have to be replaced by fresh fuel elements. In addition activation products and actinides are formed.

Fission products
Fission products are the atoms resulting from the fission of uranium atoms. Nearly all elements of the Periodic System are represented in the mixture. A substantial fraction of the fission products are highly radioactive, with half lifes varying from milliseconds to millions of years, see Tables 1-6. During the first few centuries after removal from the reactor, the radioactivity of spent sfuel is dominated by the fission products.

Actinides and minor actinides
In the IUPAC nomenclature the actinides are the heavy elements actinium and beyond in the Periodic System of the Elements. In nuclear technology the designation actinides usually denotes uranium, plutonium and other transuranium elements. Uranium occurs in nature, the transuranium elements are human-made. By capture of one neutron neptunium atoms are formed from uranium atoms, which decay to plutonium atoms. By radioactive decay and repeated neutron capture plutonium atoms are transmutated into other transuranium elements. The term *minor actinides* is usual for the radionuclides other than uranium and plutonium, such as neptunium, americium and curium.
All actinides are highly radioactive and most of them emit dangerous alpha rays and gamma rays. A number of the minor actinides exhibit spontaneous fission, causing neutron radiation and complications in an operating reactor. One of the americium isotopes has a critical mass of some 7 grams under certain conditions.
The half-lifes of the minor actinides vary from hours to millions of years (see Table 7).

Activation products
In addition to fission products and actinides a third category of radioactive atoms are generated in an operating nuclear reactor: activation products. By neutron irradiation non-radioactive construction materials become radioactive, with varying half-lifes (see Table 8).

Explanatory remarks on Tables 1-7

Tables 1-6 give the inventory of fission products (a selection) in spent fuel, at a burnup of 33 GWday/Mg and with an enrichment assay of 3.2% U-235, at moment of discharge. The figures of the inventories (last column) are from table D.1 of [KfK 1983] Q587, converted from Ci/Mg (curie per metric ton) to TBq/Mg (terabecquerel per metric ton).
For unclear reasons the authors of of KfK 1983 deleted from their tables the inventories of a number of fission products, especially Te-132, I-131, I-132, I-133 and Xe-133. This omission is remarkable, the more so because the joint activity of these radionuclides at discharge may be about ten times the activity of all other

fission products together

Some fission products have radioactive decay daughters; these are indicated by * in the tables. A radioactive decay daughter is parent of another nuclide. Due to this mechanism it is possible that radionuclides with very short half-lifes remain present in spent fuel for prolonged periods.

Table 1

Fission products which decay to near detection limits within 0.1 year (37 days) after discharge. The inventories of three radionuclides are not given in the publication of [KfK 1983] Q587. For remarks see text.

radionuclide	symbol	half-life	time frame years	decay daughter		discharge TBq/Mg
antimony-126m	^{126m}Sb	19 m	0.1	Te-126	stable	22.9
tellurium-127	^{127}Te	9.4 h	0.1	I-127	stable	3278
tellurium-129	^{129}Te	70 m	0.1	I-129	*	11 100
tellurium-132	^{132}Te	78 h	0.1	I-132	*	?
iodine-132	^{132}I	2.29 h	0.1	Xe-132	stable	?
iodine-133	^{133}I	20.8 h	0.1	Xe-133	*	?
praseodymium-144	^{144}Pr	17.3 m	0.1	Nd-144	stable	41070
praseodymium-144m	^{144m}Pr	7.2 m	0.1	Pr-144	*	488

The timeframes of these nuclides are indicated with a * in the tables. For example: Rh-103m has a half-life of 56 minutes (Table 2) and its parent nuclide Ru-103 has a half-life of 39.6 days. The short-lived nuclide Rh-103m remains present as long as Ru-103 exists. All radioactive fission products listed in Tables 1-4 decay by beta emission, often accompanied by gamma emission.

Table 2

Fission products which decay to near detection limits within 1 year after discharge. The inventories of two radionuclides are not given in the publication of [KfK 1983] Q587. For remarks see text.

radionuclide	symbol	half-life	time frame years	decay daughter		discharge TBq/Mg
ruthenium-103	^{103}Ru	39.6 d	1	Rh-103m	*	55 500
rhodium-103m	^{103m}Rh	56 m	1 *	Rh-103	stable	50 320
tellurium-129m	^{129m}Te	33.4 d	1	I-129	stable	1684
iodine-131	^{131}I	8.04 d	1	Xe-131	stable	?
xenon-133	^{133}Xe	5.29 d	1	Cs-133	stable	?
barium-140	^{140}Ba	12.8 d	1	La-140	*	57 350
lanthanum-140	^{140}La	40.2 h	1 *	Ce-140	stable	59 570
cerium-141	^{141}Ce	32.5 d	1	Pr-141	stable	54 390
praseodymium-143	^{143}Pr	13.6 d	1	Nd-143	stable	48 840
promethium-148	^{148}Pm	5.37 d	1	Sm-148	stable	6438
europium-156	^{156}Eu	15.2 d	1	Gd-156	stable	8325

Table 3

Fission products which decay to near detection limits within 10 years after discharge. For remarks see text.

radionuclide	symbol	half-life	time frame years	decay daughter		discharge TBq/Mg
strontium-89	^{89}Sr	50.5 d	10	Y-89	stable	28 897
yttrium-91	^{91}Y	58,6 d	10	Zr-91	stable	37 740
zirconium-95	^{95}Zr	65.5 d	10	Nb-95	*	54 390
niobium-95	^{95}Nb	35.1 d	10 *	Mo-95	stable	54 020
ruthenium-106	^{106}Ru	369 d	10	Rh-106m	*	18 500
rhodium-106m	^{103m}Ru	2.18 h	10 *	Pd-106	stable	20 200
silver-110	^{110}Ag	252 d	10	Cd-110	stable	4810
tin-123	^{123}Sn	129 d	10	Sb-123	stable	72.5
tellurium-127m	^{127m}Te	109 d	10	I-127	stable	429
cerium-144	^{144}Ce	284 d	10	Pr-144m	*	40 400
promethium-148m	^{148m}Pm	41.3 d	10	Sm-148	stable	1136

Table 4

Fission products which decay to near detection limits within 100 years after discharge. For remarks see text.

radionuclide	symbol	half-life	time frame years	decay daughter		discharge TBq/Mg
tritium	^{3}H	12.3 y	100	He-3	stable	18.4
krypton-85	^{85}Kr	10.7 y	100	Rb-85	stable	348
antimony-125	^{125}Sb	2,73 y	100	Te-125	stable	330
cesium-134	^{134}Cs	2.06 y	100	Ba-134	stable	15809
promethium-147	^{147}Pm	2.62 y	100	Sm-147	stable	5661
europium-154	^{154}Eu	8.6 y	100	Gd-154	stable	455
europium-155	^{155}Eu	4.8 y	100	Gd-155	stable	273

Table 5

Fission products which decay to near detection limits within 1000 years after discharge. For remarks see text.

radionuclide	symbol	half-life	time frame years	decay daughter		discharge TBq/Mg
strontium-90	^{90}Sr	29.1 y	1000	Y-90	*	2764
yttrium-90	^{90}Y	64 h	1000 *	Zr-90	stable	2908
cesium-137	^{137}Cs	30.0 y	1000	Ba-137m	*	3959
barium-137m	^{137m}Ba	2.55 m	1000 *	Ba-137	stable	3737
samarium-151	^{151}Sm	93 y	1000	Eu-151	stable	12.2

Table 6

Fission products with very long half-lifes, lt = long term. For remarks see text.

radionuclide	symbol	half-life	time frame years	decay daughter		discharge TBq/Mg
selenium-79	^{79}Se	0.065 My	lt	Br-79	stable	0,0137
zirconium-93	^{93}Zr	0.95 My	lt	Nb-93	stable	0.0662
technetium-99	^{99}Tc	0.213 My	lt	Ru-99	stable	0,485
palladium-107	^{107}Pd	6.5 My	lt	Ag-107	stable	0.00377
tin-126	^{126}Sn	0.1 My	lt	Sb-126	*	0.0201
antimony-126	^{126}Sb	12.4 d	lt *	Te-126	stable	27.6
iodine-129	^{129}I	15.9 My	lt	Xe-129	stable	0.00114
cesium-135	^{135}Cs	2.3 My	lt	Ba-135	stable	0.0130

Table 7

Actinides in spent fuel (burnup 33 GWday/Mg, initial enrichment assay of 3.2% U-235), a selection. Missing figures are indicated by n.a. (not available). Source: [KfK 1983] Q587]. According to KfK 1983 the total activity of all actinides together at discharge 1.51 EBq and the total mass of actinides is 966 kg/Mg spent fuel (excluding cladding). So the total mass of fission products is 340 kg/Mg spent fuel. SF = spontaneous fission.

radionuclide	symbol	half-life	decay mode	first decay daughter	activity at discharge TBq/Mg	content g/Mg
uranium-232	^{232}U	72 y	α SF	Th-228	0.00039	n.a.
uranium-233	^{233}U	0.16 My	α	Th-229	n.a.	0.0081
uranium-234	^{234}U	0,24 My	α	Th-230	0.034	145
uranium-235	^{235}U	704 My	α	Th-231	0.00066	8250
uranium-236	^{236}U	23.4 My	α SF	Th-232	0.0097	4050
uranium-237	^{237}U	6.75 d	β^-	Np-237	27500	n.a.
uranium-238	^{238}U	4.5 Gy	α	Th-234	0.012	943 000
neptunium-237	^{237}Np	2.14 My	α	Pa-233	0,011	426
neptunium-238	^{238}Np	2.12 d	β^-	Pu-238	11 600	n.a.
neptunium-239	^{239}Np	2.35 d	β^-	Pu-239	722000	n.a.
plutonium-236	^{236}Pu	2.85 y	α SF	U-232	0.031	0.0016
plutonium-238	^{238}Pu	87.8 y	α	U-234	83.6	132
plutonium-239	^{239}Pu	24400 y	α SF	U-235	12.4	5390
plutonium-240	^{240}Pu	6540 y	α SF	U-236	18.8	2230
plutonium-241	^{241}Pu	15 y	β^- SF	Am-241	4630	1220
plutonium-242	^{242}Pu	0.387 My	α SF	U-238	0.067	486
americium-241	^{241}Am	433 y	α SF	Np-237	4.70	37.0
americium-242m	^{242m}Am	152 y	α SF	Np-238	0.144	0.040
americium-242	^{242}Am	16 h	β^-	Cm-242	2830	n.a.
americium-243	^{243}Am	7370 y	α SF	Np-239	0.62	83.7
curium-242	^{242}Cm	163 d	α SF	Pu-238	1520	12.4
curium-243	^{243}Cm	28 y	α	Pu-239	0.62	0.32
curium-244	^{244}Cm	18 y	α SF	Pu-240	71.8	23.9
curium-245	^{245}Cm	8500 y	α	Pu-241	0.0051	0.80
curium-246	^{246}Cm	4760 y	α SF	Pu-242	0.0083	0.73

Spent fuel contains a large number of actinides, most of which are formed from uranium nuclides by neutron capture, some are decay daughters of the newly formed radionuclides. Table 7 lists only isotopes of uranium and trans-uranium elements. The decay daughters of the actinides are all radioactive.
Several actinides exhibit spontaneous fission, especially Cm-242 and Cm-244 produce high fluxes of neutron radiation.

Table 8
Important activation products in spent fuel, construction materials and coolant. Most of the radionuclides in this table decay by beta emission, some by electron capture.

radionuclide	symbol	half-life	decay daughter	
tritium	^{3}H or T	12.3 y	He-3	stable
carbon-14	^{14}C	5730 y	N-14	stable
chlorine-36	^{36}Cl	0.301 My	Ar-36	stable
calcium-45	^{45}Ca	163 d	Sc-45	stable
scandium-46	^{46}Sc	84 d	Ti-46	stable
chromium-51	^{51}Cr	28 d	V-51	stable
manganese-54	^{54}Mn	313 d	Fe-54	stable
iron-55	^{55}Fe	2.7 y	Mn-55	stable
iron-59	^{59}Fe	45 d	Co-59	stable
cobalt-58	^{58}Co	71 d	Fe-58	stable
cobalt-60	^{60}Co	5.27 y	Ni-60	stable
nickel-59	^{59}Ni	0.08 My	Co-59	stable
nickel-63	^{63}Ni	100 y	Cu-63	stable
zirconium-93	^{93}Zr	0.95 My	Nb-93	stable
zirconium-95	^{95}Zr	66 d	Nb-95	*
niobium-93m	^{93m}Nb	12 y	Nb-93	stable
niobium-94	^{94}Nb	0.02 My	Mo-94	stable
niobium-95	^{95}Nb	35 d	Mo-95	stable
molybdenum-93	^{93}Mo	3000 y	Nb-93	stable
silver-108m	^{108m}Ag	418 y	Pd-108	stable
silver-110m	^{110m}Ag	252 d	Cd-110	stable
tin-119m	^{119m}Sn	245 d	Sn-119	stable
tin-121m	^{121m}Sn	50 y	Sb-121	stable
tin-123	^{123}Sn	129 d	Sb-123	stable
antimony-124	^{124}Sb	60 d	Te-124	stable
antimony-125	^{125}Sb	2.73 y	Te-125m	*
tellurium-125m	^{125m}Te	58 d	Te-125	stable
tantalum-182	^{182}Ta	115 d	W-182	stable
tungsten-185	^{185}W	75 d	Re-185	stable
lead-205	^{205}Pb	15.3 My	Tl-205	stable

7 Radioactive decay of spent nuclear fuel

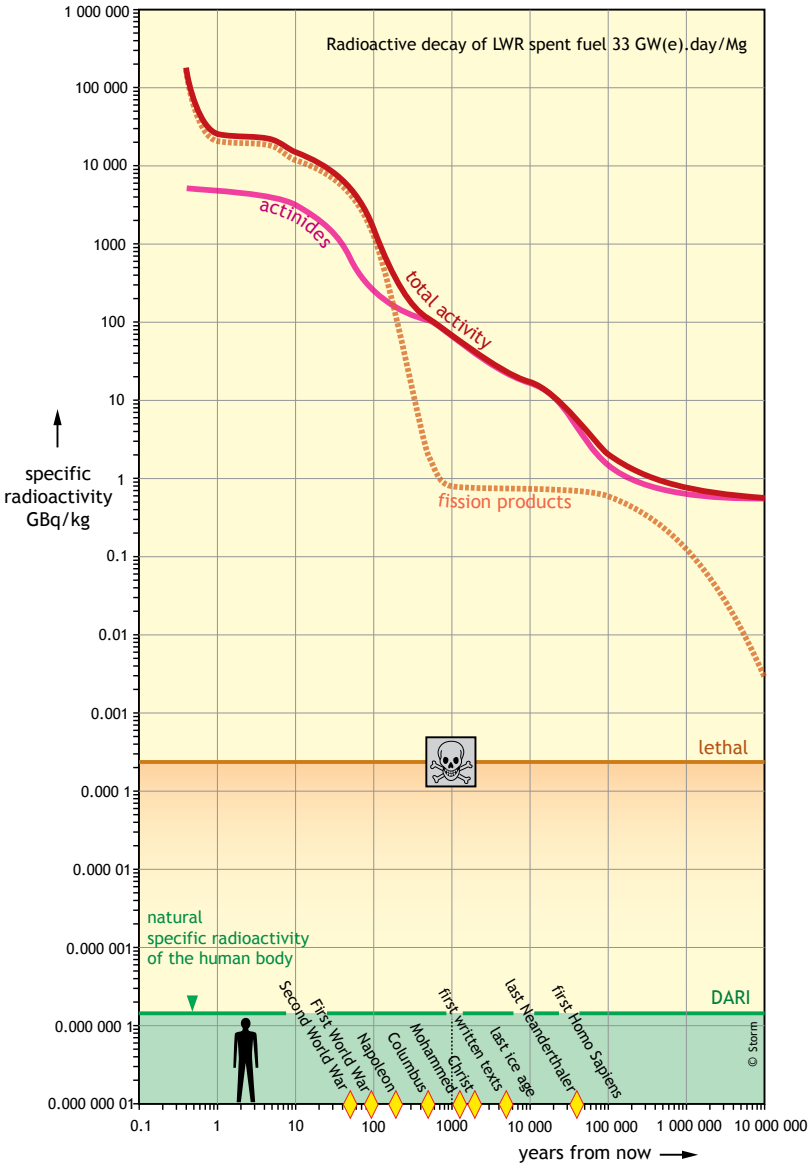

Figure 15

The specific radioactivity, in gigabecquerel per kilogram (GBq/kg), of spent fuel at a burnup of 33 GWe.day/Mg (gigawatt electric per metric tonne uranium) charged into the reactor. Nuclear fuel from current types of nuclear reactors usually has higher burnup (40-50 GWe.day/Mg) than the fuel this diagram is based on and consequently its specific radioactivity is higher. The contributions of tritium and carbon-14 are not included in these curves. Note that both axes have logarithmic scales. Each scale division denotes a factor ten. With linear time scales the horizontal axis would be about 100 kilometers long and the vertical axis some 100 million km.

On the horizontal axis a reverse historic timescale is indicated, to give an idea of the time frames involved. The green line indicates the natural radioactivity of the human body (143 Bq/kg). Sources: [Bell 1973] Q264, [Hollocher 1975] Q262, [JPL-77-69 1977] Q263, [Charpak & Garwin 2002] Q300.

The radioactivity of spent fuel at a given moment in an operating reactor is largely set by the fission products. During the fission process some thousand different nuclides are formed and a significant part of these are radioactive. During the first month after the fission process has been shut down, intentionally or by accident,

the radioactivity of spent fuel decreases sharply, due to the decay of very short-lived fission products. After the first months the radioactivity decreases slowly and is chiefly set by radionuclides with longer half-lifes. After some 300 years the radioactivity of spent fuel is chiefly set by the actinides, not to say that the very long-lived fission products are unimportant after the crossover.

From the viewpoint of health hazards it is important, among other parameters, to know which radionuclides are released from spent nuclear fuel, by whatever cause. Report m17 *Pathways of radioactive contamination* addresses the pathways along which radioactive materials from spent fuel may enter the human environment and the human body. The radioisotopic composition of the released radioactive material depends on the time period between the end of the fission process and the moment of release.

The half-lifes of the radionuclides present in used nuclear fuel vary from milliseconds to millions of years. To gain some insight into in this matter the fission products are grouped according their half-lifes in Tables 1-6. Table 1 for example lists a selection of radionuclides which have almost completely decayed within 0.1 year (37 days) after shutdown of the fission process. After 10 half-lifes only one thousandth of the original amount of a radionuclide remains, the rest has decayed to the decay daughter of the radionuclide, mostly a stable nuclide. For example, iodine-133 decays to stable xenon-133 with a half-life of 20.8 hours. Within 37 days some 42 half-lifes have gone by and about $3*10^{-13}$ of the original amount is left.

The highly hazardous actinides generally have long to very long half-lifes (see Table 7). The activation products, a minor but stil important contribution to the total radioactivity of spent fuel at the moment of discharge from the reactor, have half-lifes varying from days to thousands of years (see Table 8).

Residual heat generation

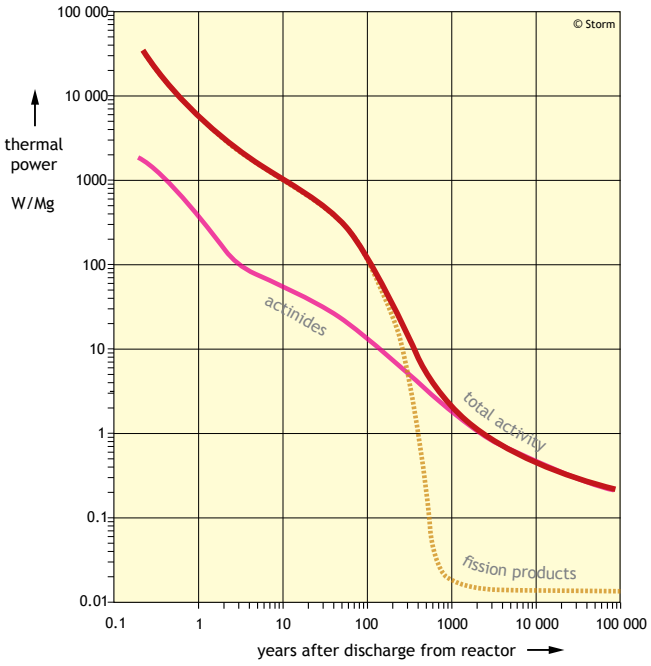

Figure 16

Residual heat generation of spent fuel (burnup B = 33 GWday/Mg) as function of cooling time. At the moment of reactor shutdown, the fuel radiates about 8 MW/Mg (not shown in the diagram). Source: [Hollocher 1975] Q262.

Note that both axes of this diagram have logarithmic scales: each division marks a factor of ten. Spent fuel from current nuclear power plants generally has a higher fuel burnup (B = 40-50 GWday/Mg) and has a about two times higher residual heat generation [IPFM 2011] Q513.

Spent fuel is extremely radioactive and generates much heat, due to the radioactive decay of its contents. The material has to be cooled in spent fuel pools for many years to prevent melting and consequently the release of the contents into the environment. The greatest part of the human-made radioactivity generated during fission is contained in the spent fuel elements.

After shutdown of the reactor the spent fuel generates so much heat, due to the radioactive decay of the fission products and actinides, that the fuel elements will melt within a very short time if not effectively cooled. The residual heat generation falls to less than 1% during the first year and slowly decays thereafter. A 100 years after removal from the reactor it is still 100-200 watt/Mg. For that reason spent fuel has to be cooled actively during many years after removal from the reactor, before it can be processed for final storage or other purposes. Even then the residual heat generation requires precautionary measures.

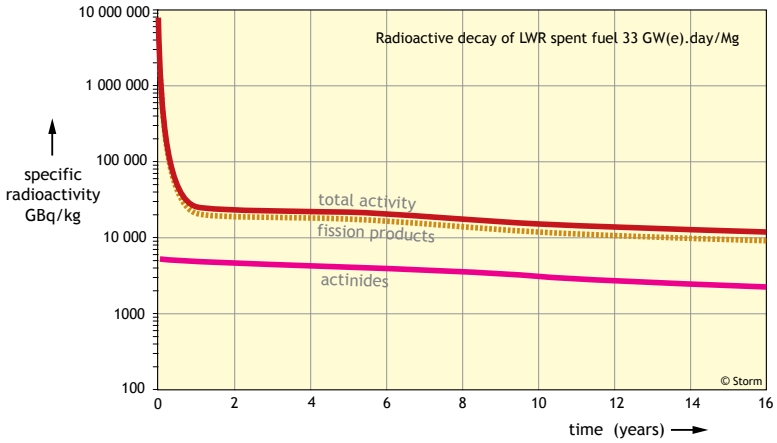

Figure 17

Radioactive decay of spent fuel as function of time. Note that the diagram has a linear horizontal (time) scale and a logarithmic vertical (activity) scale. The residual specific heat generation of spent fuel (W/kg)follows a similar curve. Source: [Bell 1973] Q264, [JPL77-69 1977] Q263.

Figure 16 represents the residual heat generation of spent fuel of relatively low burnup as function of the time; both axes, the vertical as well the horizontal, have logarithmic scales. To people who are not very used to read diagrams logarithmic scales may easily give a wrong impression of the quantities on the axes. If the horizontal axis of Figure 16 would be stretched to a linear time scale, the diagram would be 1 km long. A linear vertical scale would be 10 km high. For that reason Figure 17 is added, representing the specific radioactivity of the same type of fuel as function of the time. The time axis in Figure 17 has a linear scale, showing how slowly the specific radioactivity of spent fuel deceases with time. The specific radioactivity is a measure of the residual heat generation of spent fuel.

By far the largest part of the anthropogenic radioactivity is confined within the spent fuel elements, containing dozens of different kinds of radionuclides. The highly dangerous spent nuclear fuel is unloaded from the reactor and transported to cooling ponds, usually on the site of the nuclear power plant.
The decay of the radioactivity to levels comparable with an uncontaminated environment takes millons of years. Because of its heat generation and extremely high radioactivity spent fuel has to be cooled for decades, in order to avoid melting, hydrogen explosions and consequently release of its contents, as happened in Fukushima.

8 Spent fuel management

Spent fuel inventories

A typical modern reactor has a capacity of about 1 GWe (1,000 Megawatts electric). According to the International Atomic Energy Agency, there exist today 331 GWe of LWRs, 23 GWe of PHWRs, and 19 GWe of graphite-moderated reactors. Almost all the reactors now under construction are LWRs, and indeed most are PWRs.
The amount of spent fuel discharged from a nuclear power plant per unit delivered electricity depends upon the fuel burnup, i.e., the thermal energy (heat) generated per unit mass of fuel. Table 9 shows the approximate amount of spent fuel that would be discharged per year from a 1 GWe reactor of the three most common reactor types.
As of the end of 2009, there were about 240,000 metric tons (as heavy metal) of spent fuel in storage worldwide, most of it at reactor sites. About 90% was in storage ponds, the balance was
in dry-cask storage. The annual spent fuel generated is approximately 10,500 tons of heavy metal per year, with roughly 8500 tons of heavy metal going into long term storage and about 2000 tons of heavy metal allocated for reprocessing but much of it in interim storage [IPFM 2011] Q513.

Table 9
Annual discharge of spent fuel for three common reactor types. This assumes a reactor of 1 GWe operating at 90% capacity. GWd/tHM is the amount of thermal energy (heat) in gigawatt-days released per metric ton of heavy metal (HM) in the fuel. Source: [IPFM 2011] Q513.

reactor type	typical burn-up (GWd/tHM)	annual discharge of spent fuel (Mg)
LWR (light-water moderated)	50	20
CANDU (heavy-water moderated)	7	140
RBMK (graphite moderated)	15	65

The most systematic reporting on spent fuel inventories is done by the national reports required under the Joint Convention on the Safety of Spent Fuel Management and the Safety of Radioactive Waste Management. The spent fuel inventories for the countries covered in this study, which account for over 80% of global nuclear capacity, are shown in Table 9, with totals for France and Japan reported from other sources.
The U.S. has by far the largest holding of spent fuel. As of the end of 2010, the total U.S. stockpile of spent power-reactor fuel was 64,500 Mg, including 15,350 Mg in dry casks [IPFM 2011].

The composition, heat output and radioactivity per ton of heavy metal of the spent fuel depend upon the burn-up. For LWR spent fuel with a burnup of 50 GWd/MgHM, 1 Mg spent fuel consists of about 934 kg uranium (of which about 0.8% U-235), 52 kg fission products, 12 kg plutonium (corresponding with 1.5 weapon equivalents per Mg of fuel), and 2 kg minor transuranic elements (neptunium, americium, curium).

Table 10

Spent fuel inventories in cooling ponds and dry-cask storage at the end of 2007 for the 10 countries in the present study. Source: [IPFM 2011] Q513

Country	Spent Fuel Inventory (Mg of heavy metal) end of 2007	Spent Fuel Policy
Canada	37,300	Direct disposal
Finland	1,600	Direct disposal
France	13,500	Reprocessing
Germany	5,850	Direct disposal (now)
Japan	19,000	Reprocessing
Russia	13,000	Some reprocessing
South Korea	10,900	Storage, disposal undecided
Sweden	5,400	Direct disposal
United Kingdom	5,850	Reprocessing but future unclear
United States	61,000	Direct disposal
total	173400	

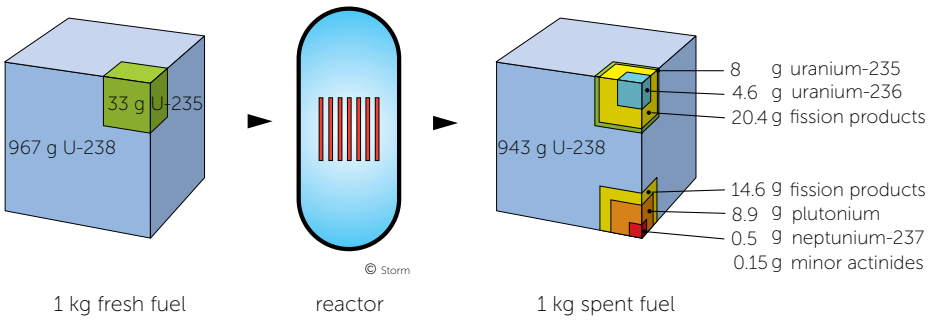

Figure 18

Composition of fresh and spent nuclear fuel. Fresh nuclear fuel consists of very pure uranium, enriched in the fissile uranium-235 isotope. During operation of the reactor a part of the U-235 nuclides are fissioned. a part is converted into non-fissile U-236 and a part has not fissioned when the fission process is no longer sustainable and the fuel has to be removed from the reactor. By neutron capture a small part of the non-fissile U-238 isotopes are converted into fissile and non-fissile plutonium isotopes. A part of the formed plutonium is fissioned and so contributes to the energy production. Another part of the plutonium is converted into the minor actinides: nuclides with a higher atomic number than plutonium. The radioactivity of 1 kg spent fuel is a billion times higher than of 1 kg fresh fuel.
In reality the constituents of fresh and spent nuclear fuel are mixed on atomic scale, so it is not possible to cut out a few pieces from spent fuel to obtain all fission products, plutonium and minor actinides in separate blocks. Separation of these materials requires a complicated sequence of processes, called reprocessing.

Spent fuel management options

The bulk of the human-made radioactivity is contained in the spent fuel leaving the nuclear reactor. The nuclear industry suggests three options to deal with spent fuel:

1 Direct disposal as radioactive waste
2 Reprocessing and vitrification
3 Partitioning and transmutation (P&T).

According to a popular view within the nuclear industry the latter two technical concepts, vitrification and P&T, could reduce the high-level waste problem to a routine job, nothing to worry about, see for example [MacKay 2009] Q399. Seemingly the proponents of options 2 and 3 are fooled by the appearance of Figure 18: only a small mass fraction of spent fuel is dangereously radioactive, overlooking the fact that all radionuclides are mixed throughout the mass of spent fuel at atomic scale.
Both concepts may look plausible at a first glance. However, on closer examination these concepts prove to be unfeasible as radioactive waste reduction strategies, because they are based on fallacies and ignorance of the Second Law of thermodynamics. Even if feasible the energy consumption of concepts 2 and 3 would be prohibitive.

Option 1, permanent storage in a geologic repository without any treatment other than packing in durable containers, is the least dangerous way to isolate the radioactivity in spent fuel from the human environment. The next section briefly addresses technical aspects of direct disposal of spent fuel. By leaving the spent fuel elements intact, the volume containing the radioactivity remains minimal and the contents are in least unstable form. Consequently the chances of dispersion of the radioactivity into the environment remain minimal. The less activities involving the spent fuel, the less chances of accidents and releases of the radioactive contents. In addition, the energy consumption of this option is by far less than of the other options.

Option 1 is decribed in detail in reports m32 *Geologic repositories* and m40 *Radioactive waste management, future CO_2 emissions*. The two other options are discussed separately in the reports m30 *Vitrification of nuclear waste* and m16 *Partitioning and transmutation*. Reprocessing technology is crucial in the concepts of vitrification and P&T. Technical and environmetal aspects of reprocessing are addressed in report m20 *Reprocessing of spent nuclear fuel* and its inherent limitations are addressed in report m38 *Nuclear power and the Second Law*.

The relation between half-life and specific activity of a radionuclede cab be calculated by the following equation.

$$-\frac{dn}{dt} = \frac{N_A \ln 2}{M\ t_{1/2}\ c} = \frac{1.323 \cdot 10^{16}}{M\ t_{1/2}}$$

$\frac{dn}{dt}$	= specific activity	Bq g^{-1}
N_A	= constant of Avogadro = $6.022 \cdot 10^{23}$	mol^{-1}
M	= molar mass radionuclide	g mol^{-1}
$t_{1/2}$	= half-life of the radionuclide	year
c	= constant = $365.25 \cdot 24 \cdot 3600$	sec $year^{-1}$

9 Radioactive effluents and discharges

Data on the radioactive effuents of nuclear installations are scarce in the open literature. Below some tables

Table 11 Activity of some radionuclides in mill tailings
TBq per GW_e.a, LWR once through

	activity in TBq
U-238	1.3
U-234	1.3
Th-230	25.2
Ra-226	26.6
Pb-210	26.6
total	81.0

Source: INFCE-7 1980 [Q277]

Table 12 Discharges in the effluents of the front end of a LWR chain, in TBq/GW(e).a
Source: Pigford et al. 1973 [Q112]

	gaseous	liquid	remarks
mining + milling	2.101	0.125	sum U, Th-230, Ra-226, Rn-222
mill tailings			in solids: 4.07 TBq/GW(e).a
conversion	0.0023	0.011	sum U, Th-230, Ra-226
enrichment	0.0011	0.000074	U
depleted uranium			2.08 TBq/GW(e).a, 0.2% U-235
fuel element fabrication	0.0007	0.000007	U
total front end	2.105	0.136	

Table 13 Approximate discharge limits of radionuclides in liquid effluent to rivers and sea of one LWR nuclear power plant in Europe in 2000.
Source: OSPAR 2002 [Q236]

radionuclide	discharge limits TBq/yr	remarks
tritium	20 - 70	
other nuclides	0.02 - 0.4	not specified
gross alpha	0.0002	only given for Borssele (NL), possibly in other countries similar limits
gross beta (excl tritium)	n.a.	not published for LWR

Table 14 Discharges of some radionuclides in the effluents of PWR's, in TBq/GW(e).a
Sources: NEA 1980 [Q75], Pigford et al. 1973 [Q112] and various NCRP reports (see references Q251, Q256, Q259 and Q266).
n.m. = not mentioned

	NEA 1980	Pigford 1973	NCRP	
H-3	37	18.5	31	NCRP-62 1995
C-14	0.3 - 0.4	n.m.	0.37 - 0.52	NCRP-81 1993
Kr-85	10	–	<110	NCRP-44 1975
Kr + Xe	–	259	–	
I-131	n.m.	0.030	–	
I-129	n.m.	n.m.	0.000018 ?	NCRP-75 1983 *
other nuclides	n.m.	0.185	–	

* Assumed 0.1% of fission products escape from leaking fuel pins.

Table 15 Discharge limits of reprocessing plants at La Hague.
n.a. = not available. Source: Malherbe 1991 [Q17]

radionuclide	gaseous effluent TBq/yr	liquid effluent TBq/yr
tritium	2200	37000
Kr	480000	–
halogens	0.11	n.a.
aerosols	0.074	–
total alpha	n.a	1.7
total beta	n.a	1700
of which Cs-137 + Sr-90		220

10 Final disposal of other radioactive wastes

Apart from spent fuel the nuclear energy system produces large volumes of other categories of radioactive waste, that do not produce heat at an appreciable rate. As there are no melting risks, these wastes can be stacked in large quantities in underground caverns. As the waste containers contain equally hazardous radionuclides as spent fuel, albeit in lower concentrations, the caverns should also be backfilled with bentonite to retard the migration of radionuclides from leaking containers.
In a number of countries this approach is being considered, but it is nowhere practicised. A design example of such an underground storage facility is the Swedish SFR concept desctibed in reports m32 *Geologic repositories* and m40 *Radioactive waste management, future CO_2 emissions*.

Operational waste

All activities and industrial processes involving radioactive materials generate radioactive waste. Especially reprocessing plants are extremely polluting.
Usually this waste is packed in drums or concrete containers and stored in temporary storage facilities. It may contain all kinds of hazardous radionuclides, fission products, actinides and activation products, in relatively low concentrations, but nonetheless hazardous.
Ultimately the waste containers have be stored in a geologic repository.

Mining waste

Uranium mining is a source of contamination of vast areas with weakly radioactive but nevertheless hazardous dust, for reason of its content of dangerous alpha-emitting radionuclides. At the mining site the groundwater is contaminated with radioactive and chemically toxic substances, a problem expanding spatially with time.
The volumes of the radioactive waste resulting from uranium mining are enormous. These wastes have to be immobilised at the mining site. The character of the mining wastes and the way to minimise the impact of these wastes on the environment are discussed in reports m26 *Uranium mining + milling* and m41 *Uranium mine rehabilitation*.

Decommissioning and dismantling waste

As a result of the intense neutron radiation during the fission process highly radioactive activation products are formed in the construction materials of a nuclear reactor and its associated appendages. Due to contamination with radioactive corrosion products and radioactive materials from leaking fuel pins the reactor vessel and the primary cooling circuit become radioactive to a much wider extent. After closedown of a power plant the so-called nuclear island has to be decommissioned and dismantled. These activities generate very large volumes and masses of radioactive waste: tens of thousands of tonnes and tens of thousands of cubic meters, see report m04 *Decommissioning and dismantling*.
The specific radioactivity of these wastes (measured per tonne waste), containing not only activation products, but also fission products and actinides as a result of contamination during the operational lifetime, is lower than that of spent nuclear fuel, but the total content of radioactivity is very large and long-lived. The waste is dangerous to man even after millions of years. The decommissioning and dismantling wastes have to be packed in appropiate containers and disposed of in a geologic repository, despite its large volumes and masses.

Dilution and immobilisation

Immobilisation of human-made radioactivity in the 'natural way', like the radioactive elements in uranium ore as suggested by WNA 2012b [Q541], is not possible for two reasons:

- Quantitatively because the quantities of radioactivity are ten million times (after 10 years of cooling) of the original natural amounts. For each tonne of uranium ore used in the nuclear chain ten million tonnes of artificial waste rock would be required. This is not to say that uranium ore in itself is harmless, on the contrary, exposure to it has deleterious health effects. Untouched uranium ore bodies deeply underground, mostly billions of years old, have the least interaction with the biosphere.

- Qualitatively because the radioactive waste stream from the reactor contains dozens of different kinds of radionuclides, almost all elements of the Periodic Table are represented, each having its own physical and chemical properties. Some radionuclides are volatile, others do not form stable and/or insoluble chemical compounds. Only a limited number of the radionuclides present in the mix could be chemically converted into stable compounds.

Just one option: isolation from the biosphere

The two advanced waste management options, vitrification and partitioning & transmutation, proposed by the nuclear industry are based on technical concepts only possible in cyberspace. Contrary to the promises of the nuclear industry both options would cause massive releases of radionuclides into the enviroment and would greatly increase the risks of large=acale dispersion of radioactivity.
There is just one survival strategy to deal with the radioactivity set free by nuclear power. As radioactivity cannot be destroyed, nor can be made harmless to man, it must be kept out of the human environment for at least a million of years. In other words: the radioactive waste has to be physically immobilised and isolated permanently from the biosphere, as soon as possible and effective as possible. See report m32 *Geologic repositories*..

This view applies to all kinds of radioactive waste, however 'low-level' it were designated by the nuclear industry. Little is known about the, always deleterious, health effects of exposure to radioactive contaminants. Still less is known about the health effects of chronic exposure to many different radionuclides simultaneously. Empirical evidence, usually neglected by the nuclear industry, proves that prolonged exposure even to very low doses of a number of radionuclides has serious health effects, as pointed out in report m11 *Health effects of radioactivity*.
The fact that the health effects of exposure to 'low' doses of radioactivity have long latency periods, should be no reason to down play the hazards of radioactivity. This issue will be discussed in more detail in report m05 *Downplaying and denial of health effects*.

If the radioactivity mobilised and generated by human activities is not effectively isolated from the human environment, one can be sure that permanent exposure to increasing numbers and amounts of radioactive materials will occur in vast and ever expanding densily inhabited areas, as a consequence of natural deteriorating mechanisms which follow from the Second Law of thermodynamics. Dispersion of radioactivity never stops and can only increase with time. This problem is aggravated by human intervention at above-ground storage sites, of which the knowledge of its dangerous radioactive contents will be lost within decades. This observation is already actual.

The above observations point to the sole remaining strategy to deal with radioactive wastes: effective and permanent isolation of the radionuclides from the human environment by means of human-made and geologic barriers in a *deep geologic repository* such as described in report m32 *Geologic repositories*. Nowhere in the world such a repository is available.

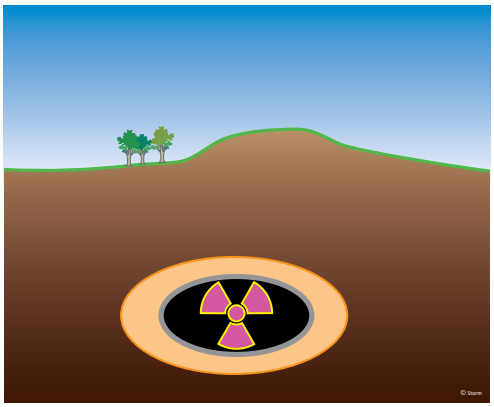

Figure 19

Symbolic representation of a deep geologic repository. The purpose is to isolate the radionuclides from the biosphere for geologically long periods.

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