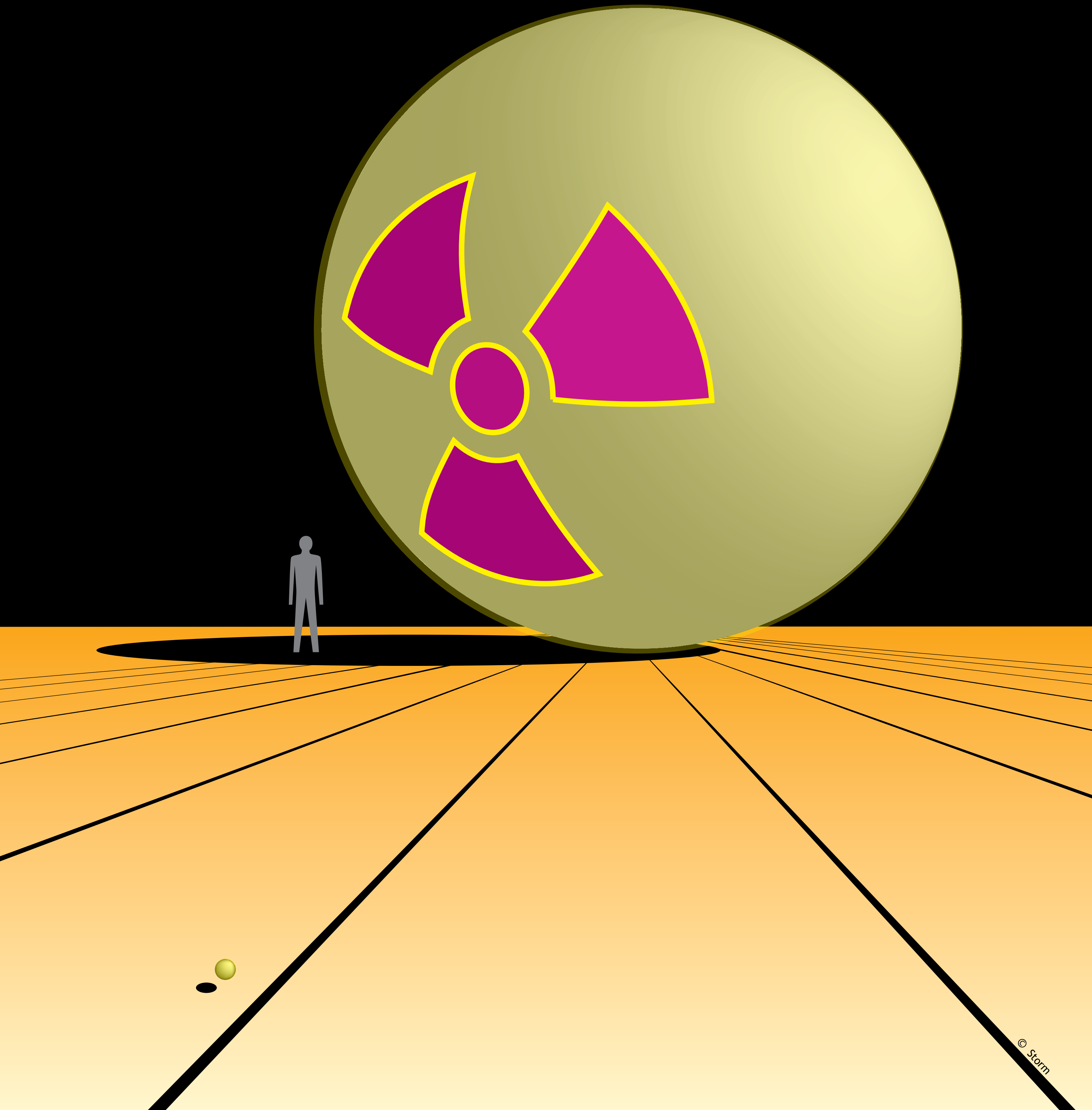
IN CAUDA VENENUM

HEALTH HAZARDS OF NUCLEAR POWER



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HEALTH HAZARDS OF NUCLEAR POWER

ANNEXES

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Abstract

This study starts with a physical assessment of the quantities of the radioactivity being generated and mobilied by the entire system of related industrial processes making civilian nuclear power possible. It assesses the actual and potential exposure of the public to human-made radioactivity, and it discusses empirical evidence of harmful health effects of these exposures. The biomedical effects of radionuclides in the human body are briefly discussed.

Furthermore this study analyses the mechanisms which may cause the uncontrolled dispersion of very large amounts of radioactivity into the environment. The study explains some consequences of a basic law of nature (Second Law) for the health risks of nuclear power now and in the future. Misconceptions, uncertainties and unknowns of the nuclear safety issue are addressed. Risk enhancing factors are discussed, along with the consequences of the present economic paradigm for the health risks of nuclear power at this moment and in the future.

The hazards of nuclear power just do not stop at the reactor: what happens and what will happen with the human-made radioactivity? In causa venenum.

Acknowledgements

The author would like to thank Ian Fairlie for reviewing the radiological part of this report and for his suggestions, Angelo Baracca for his suggestions, and Stephen Thomas and John Busby for their comments. The author notes that this report does not necessarily reflect their opinion.

In cauda venenum

A Latin phrase from ancient Rome, meaning: the poison is in the tail. Using the metaphor of a scorpion, this can be said of a story or development that proceeds gently, but turns vicious towards the end.

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ANNEX A

Radioactivity

A.1 Radioactivity, some basics

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.

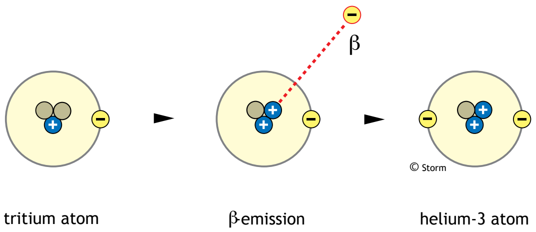


Figure A1.
Radioactive decay of tritium.Tritium, symbols T, ^{3}H or H-3, is a heavy isotope of hydrogen, with one proton and two neutrons in the nucleus. When a tritium atom decays, it emits a beta particle (an electron) at high speed. After decay the nucleus contains two protons and one neutron, the 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

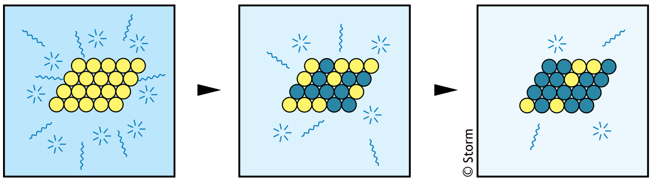
Nuclear radiation is often called ionising radiation, because it strongly interacts with matter forming ions. Ionising radiation is harmful to living organisms, for it destroys biomolecules. Alpha and beta radiation can be blocked by thick paper respectively aluminum foil, so these rays may seem not 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 A2.
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.



time t = 0 after 1 half-life after 2 half-lifes

© Storm

Figure A3.
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.

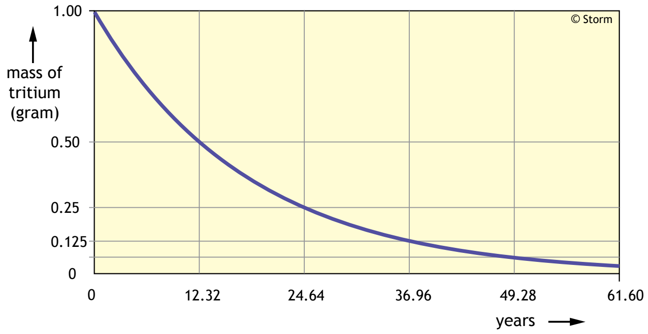


Figure A4.
The mass of a given amount of tritium as function of the time. After one half-life half of the initial number of tritium atoms has decayed into helium-3 atoms. During the second half-life period half of the remaining tritium atoms decay. Radioactive decay is a stochastic process.

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 man-made radioactivity is a billion times the radioactivity of the fresh uranium entering the reactor.

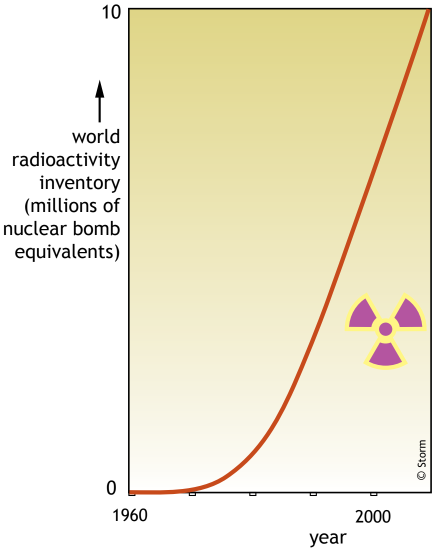


Figure A5.
All man-made radioactivity still exists in a mobile form in the human environment. The world inventory of man-made radioactivity passed the 10 million nuclear bomb equivalent mark in 2010 and is rising nearly linearly with 370 000 nuclear bomb equivalents a year.

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.

A.2 Nuclear radiation

Radioactive atoms (radionuclides) have unstable nuclei, which turn spontaneously into stable nuclei. This process is coined radioactive decay and cannot be controlled by man. Radioactive decay is accompanied by the emission of radiation from the nucleus. There are three main types of nuclear radiation: α (alpha), β (beta) and γ (gamma) rays.

Alpha rays consist of the bare nuclei of helium-4, which are ejected from the decaying nucleus with high energy. Due to their electric charge and high speed, the interaction of alpha particles with normal matter is very strong. As a result of the strong interaction the range of α-rays within solid matter is very short: they are blocked by a heavy sheet of paper. Consequently α-rays do not penetrate deeply into the skin.

Beta rays consist of electrons which are ejected at high speed from the decaying nucleus. The interaction of β-rays with matter is less strong than of α-rays and consequently their range is longer. Beta particles are stopped by 1 mm aluminum.

Gamma radiation is a kind of electromagnetic radiation, more energetic than X-rays. Gamma emission occurs usually in combination with alpha- or beta emission. Gamma rays have a weaker interaction with matter than α- and β-rays and are very penetrating, they can pass through thick layers of metal and concrete.

During interaction with the atoms and molecules of normal matter the majority of the energy of the nuclear radiation is given off to electrons, which are knocked loose from the atoms. The energy of these secondary electrons is 1-2- eV (electronvolt), about the energy range of visible and ultraviolet light, is enough to cause chemical reactions. An atom which looses one or more electrons becomes a positively charged ion, a chemically very reactive species. For reason of the generation of ions in matter, nuclear radiation is often called ionising radiation.

Alpha- and beta-rays from a source outside of the body do not penetrate far into the skin. If however radionuclides are absorbed by the body, e.g. by inhalation or ingestion, the emitted rays pass through unshielded tissue and cause biological damage. Alpha particles are more devastating than beta- and gamma-rays, due to their high electric charge and higher energy. On its short path through living tissue an α-particle from a decaying uranium atom generates about 16000 secondary electrons. Each of these electrons can initiate a chemical reaction. For reason of the high number of secondary electrons and the high LET value (Linear Energy Transfer = the amount of energy dissipated per unit path length) α-emitters are classified as the most harmful type of radionuclides in living organisms.

Spent fuel contains relatively large amounts of α-emitters in the form of actinides. In addition to the dangerous α-emission, several of the actinides, e.g. americium and curium, exhibit spontaneous fission. As a result spent fuel also emits neutrons, which are biologically very dangerous. Neutrons are not ionising directly, they don’t have an electric charge, but interact with organic molecules via activation reactions. The penetrating power of neutrons is very high. The biologically harmful effects of fast neutrons are assumed to be ten times as high as those of gamma-radiation.

A.3 Radioactive materials

Spent nuclear fuel

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 atoms. During the fission process highly radioactive fission products and actinides are formed. When the fissile content of the nuclear fuel decreases below a certain level the fission process cannot sustain itself and the fuel is removed from the reactor as spent nuclear fuel. 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. More than 90% of the man-made radioactivity generated during fission is contained in the spent fuel elements.

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 seconds to millions of years.
During the first four centuries after removal from the reactor, the radioactivity of spent sfuel is mainly determined by the fission products.

Actinides and minor actinides

Actinides are radionuclides formed from uranium atoms by neutron capture and have a higher atom number (= number of protons in the nucleus) than uranium. First neptunium atoms are formed, which quickly decay to plutonium atoms. By repeated neutron capture the plutonium atoms are transmutated into still heavier atoms. These elements, including plutonium, are often called the actinides and do not occur in nature. The minor actinides are the radionuclides beyond plutonium, such as americium, curium and californium. All actinides are highly radioactive and most of them emit dangerous alpha 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 a few decades to millions of years.

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, often with very long half-lifes.

A.4 Radioactive decay of human-made radioactivity

Spent fuel contains fission products, newly-formed actinides, activation products and unfissioned uranium. During the first year after shutdown of the fission process, the radioactivity per kilogram falls with a factor of hundred, after that much more slowly. The decay of the radioactivity of spent fuel is represented for a period of ten million years in Figure A6. To bring the time intervals within human perspective, some important dates are given, looking back in time as far as the decay will take into the future.

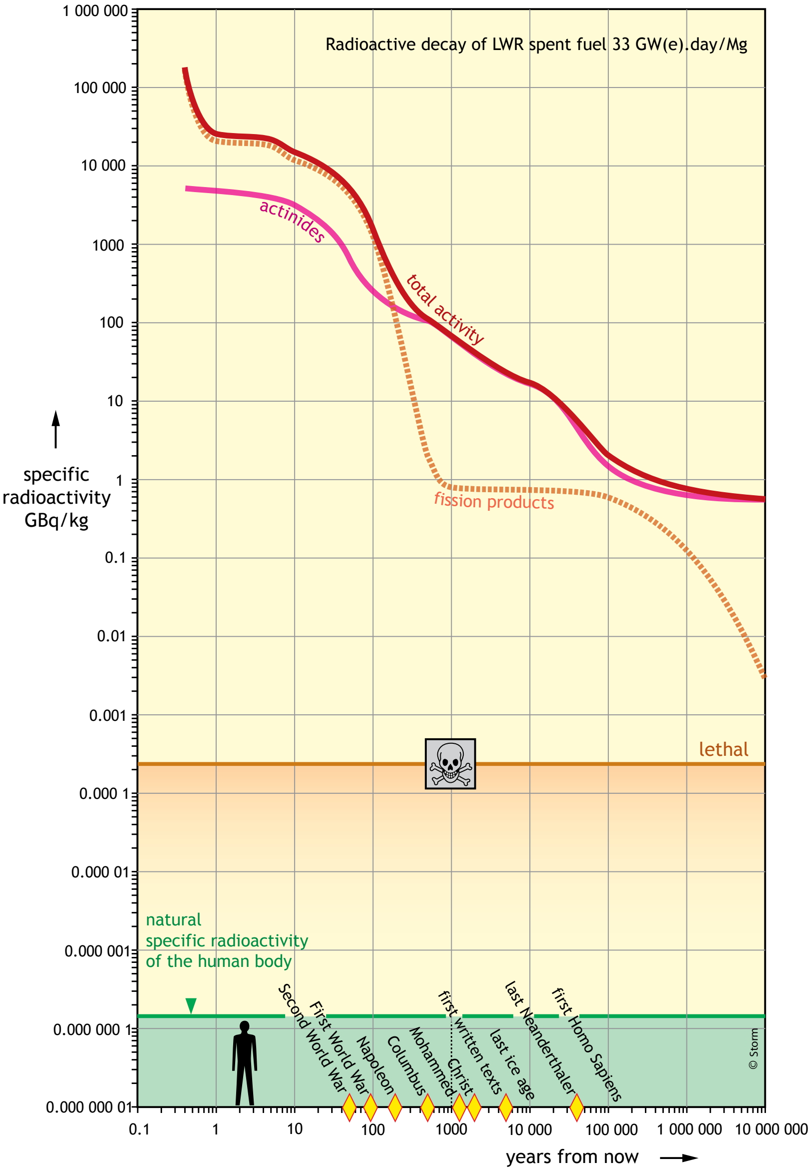


Figure A6

The specific radioactivity, in gigabecquerel per kilogram (GBq/kg), of spent fuel. Nuclear fuel from the new types of nuclear reactors has considerably higher burnup than the fuel this diagram is based on and so is its specific radioactivity. The contributions of activation products, including tritium, carbon-14, are not included in these curves. Note that both axes have logarithmic scales. Each scale division denotes a factor ten. With a linear time scale the horizontal axis would be about 100 kilometers long.

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). For more details see text. Sources: Bell 1973 Q264 2], [3] Hollocher 1975 Q262, [4] JPL-77-69 1977 Q263, [17] Charpak & Garwin 2002 Q300.

Table 1

Inventory of fission products (selection) in spent fuel, 33 GWday/Mg, 3.2% U-235, at moment of discharge. The figures of the inventories (last column) are from KfK 1983 Q587 (table D.1), converted from Ci/Mg (curie per metric ton) into TBq/Mg (terabecquerel per metric ton). For unclear reasons the inventories of a number of fission products are deleted from the tables in the original publication.

r = radioactive decay daughter, parent of another nuclide.

The meaning of the fifth column (‘tme frame’) will be explained in the text.

radionuclide	symbol		half-life	time frame years	decay daughter		discharge TBq/Mg
tritium	T, H-3	^{3}H	12.3 y	100	He-3	stable	18.4
selenium-79	Se-79	^{79}Se	65 000 y	lt	Br-79	stable	0,0137
krypton-85	Kr-85	^{85}Kr	10.7 y	100	Rb-85	stable	348
strontium-89	Sr-89	^{89}Sr	50.5 d	10	Y-89	stable	28 897
strontium-90	Sr-90	^{90}Sr	29.1 y	1000	Y-90	r	2764
yttrium-90	Y-90	^{90}Y	64 h	1000 *	Zr-90	stable	2908
yttrium-91	Y-91	^{91}Y	58,6 d	10	Zr-91	stable	37 740
zirconium-93	Zr-93	^{93}Zr	0.95 My	lt	Nb-93	stable	0.0662
zirconium-95	Zr-95	^{95}Zr	65.5 d	10	Nb-95	r	54 390
niobium-95	Nb-95	^{95}Nb	35.1 d	10 *	Mo-95	stable	54 020
technetium-99	Tc-99	^{99}Tc	0.213 My	lt	Ru-99	stable	0,485
ruthenium-103	Ru-103	^{103}Ru	39.6 d	1	Rh-103m	r	55 500
ruthenium-106	Ru-106	^{106}Ru	369 d	10	Rh-106m	r	18 500
rhodium-103m	Rh-103m	^{103m}Rh	56 m	1 *	Rh-103	stable	50 320
rhodium-106m	Rh-106	^{103m}Ru	2.18 h	10 *	Pd-106	stable	20 200
palladium-107	Pd-107	^{107}Pd	6.5 My	lt	Ag-107	stable	0.00377
silver-110	Ag-110	^{110}Ag	252 d	10	Cd-110	stable	4810
tin-123	Sn-123	^{123}Sn	129 d	10	Sb-123	stable	72.5
tin-126	Sn-126	^{126}Sn	0.1 My	lt	Sb-126	r	0.0201
antimony-125	Sb-125	^{125}Sb	2,73 y	100	Te-125	stable	330
antimony-126	Sb-126	^{126}Sb	12.4 d	lt *	Te-126	stable	27.6
antimony-126m	Sb-126m	^{126m}Sb	19 m	0.1	Te-126	stable	22.9
tellurium-127	Te-127	^{127}Te	9.4 h	0.1	I-127	stable	3278
tellurium-127m	Te-127m	^{127m}Te	109 d	10	I-127	stable	429
tellurium-129	Te-129	^{129}Te	70 m	0.1	I-129	r	11 100
tellurium-129m	Te-129m	^{129m}Te	33.4 d	1	I-129	r	1684
tellurium-132	Te-132	^{132}Te	78 h	0.1	I-132	r	?
iodine-129	I-129	^{129}I	15.9 My	lt	Xe-129	stable	0.00114
iodine-131	I-131	^{131}I	8.04 d	1	Xe-131	stable	?
iodine-132	I-132	^{132}I	2.29 h	0.1	Xe-132	stable	?
iodine-133	I-133	^{133}I	20.8 h	0.1	Xe-133	r	?
xenon-133	Xe-133	^{133}Xe	5.29 d	1	Cs-133	stable	?
cesium-134	Cs-134	^{134}Cs	2.06 y	100	Ba-134	stable	15809
cesium-135	Cs-135	^{135}Cs	2.3 My	lt	Ba-135	stable	0.0130
cesium-137	Cs-137	^{137}Cs	30.0 y	1000	Ba-137m	r	3959
barium-137m	Ba-137m	^{137m}Ba	2.55 m	1000 *	Ba-137	stable	3737
barium-140	Ba-140	^{140}Ba	12.8 d	1	La-140	r	57 350
lanthanum-140	La-140	^{140}La	40.2 h	1 *	Ce-140	stable	59 570
cerium-141	Ce-141	^{141}Ce	32.5 d	1	Pr-141	stable	54 390
cerium-144	Ce-144	^{144}Ce	284 d	10	Pr-144m	r	40 400
praseodymium-143	Pr-143	^{143}Pr	13.6 d	1	Nd-143	stable	48840
praseodymium-144	Pr-144	^{144}Pr	17.3 m	0.1	Nd-144	stable	41070
praseodymium-144m	Pr-144m	^{144m}Pr	7.2 m	0.1	Pr-144	r	488
promethium-147	Pm-147	^{147}Pm	2.62 y	100	Sm-147	stable	5661
promethium-148	Pm-148	^{148}Pm	5.37 d	1	Sm-148	stable	6438
promethium-148m	Pm-148m	^{148m}Pm	41.3 d	10	Sm-148	stable	1136
samarium-151	Sm-151	^{151}Sm	93 y	1000	Eu-151	stable	12.2

europium-154	Eu-154	^{154}Eu	8.6 y	100	Gd-154	stable	455
europium-155	Eu-155	^{155}Eu	4.8 y	100	Gd-155	stable	273
europium-156	Eu-156	^{156}Eu	15.2 d	1	Gd-156	stable	8325

* These short-lived radionuclides are constantly formed by decay of other radionuclides and consequently remain present as long as their parent nuclides are present.

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

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

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

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

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

Table 4

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

Table 2

Important activation products in spent fuel.

radionuclide	symbol		half-life	decay daughter	
carbon-14	C-14	^{14}C	5730 y	N-14	stable
manganese-54	Mn-54	^{54}Mn	313 d	Fe-54	stable
iron-55	Fe-55	^{55}Fe	2.7 y	Mn-55	stable
cobalt-60	Co-60	^{60}Co	5.27 y	Ni-60	stable
nickel-63	Ni-63	^{63}Ni	100 y	Cu-63	stable
silver-110m	Ag-110m	^{110m}Ag	252 d	Cd-110	stable

The numbers on the vertical scale of Figure A6 are in gigabecquerel per kilogram mass (GBq/kg, 1 Bq = 1 desintegration per second). This measure of specific radioactivity may have little meaning without a reference value. The obvious standard is the human body itself, which has a natural specfic radioactivity of about 143 Bq/kg, mainly due to potassium-40 (86 Bq/kg) and carbon-14 (57 Bq/kg), two naturally occurring radioisotopes. The average lethal amount of radioactivity in the human body is roughly 25000 times the natural activity [17] Charpak & Garwin 2002 Q300. which is indicated in Figure A6, but this amount strongly depends on the types of radionuclides.

The curves of Figure A6 do not include the activities of tritium and carbon-14, nor the activity of the radionuclides in the cladding hulls of the fuel. The contributions of these nuclides may be significant after a cooling period of 10 years.

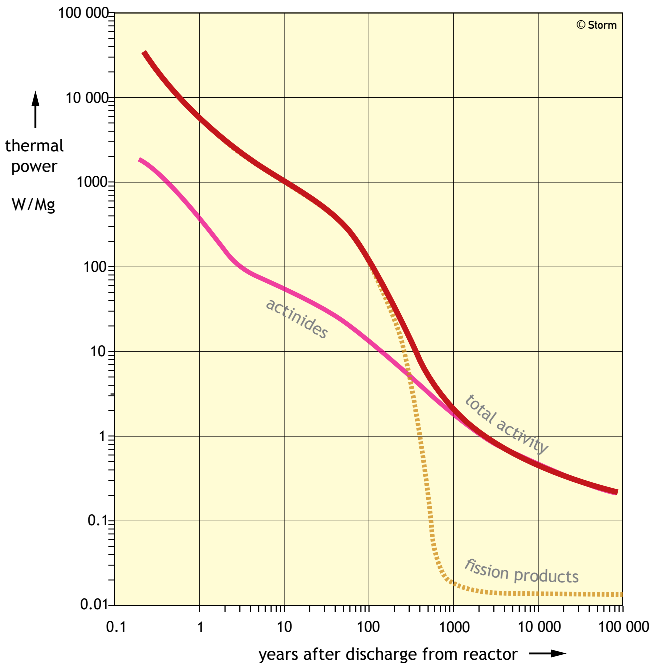


Figure A7

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). Spent fuel from modern nuclear power plants generally has a higher fuel burnup (*B* = 40-50 GWday/Mg) and consequently has a higher residual heat generation. Source: Hollocher 1975 [Q262]. Note that both axes of this diagram have logarithmic scales: each division marks a factor of ten. 1 Mg = 1 megagram = 1 metric tonne.

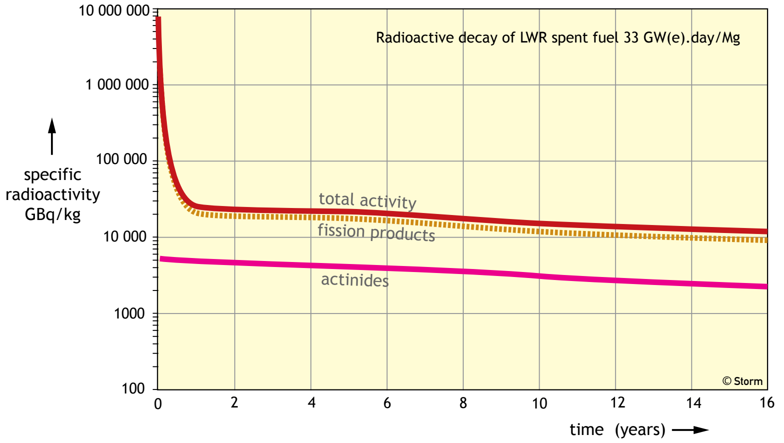


Figure A8

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].

A.5 Some quantities and units

Activity, unit becquerel (Bq)

The SI unit of radioactivity is the *becquerel*, symbol Bq:
1 Bq = 1 desintegration per second.

The old unit of activity, still commonly used, is the *curie* (Ci):
1 Ci = $3,7 \bullet 10^{10}$ desintegrations per second, or:
1 Ci = 37 GBq
The curie had been deduced from the observation that 1 gram of radium-226 produces about $3,7 \bullet 10^{10}$ desintegrations per second.

Absorbed dosis *D*, unit gray (Gy)

The present dose-effect models assume that the damage inflicted to living tissue by nuclear radiation depends on the amount of energy transferred to the living matter. The more energy, the more damage.
The *gray* (symbol Gy) is de SI unit of exposure, officially: the unit of the absorbed dose *D*. The gray is a measure of the amount of absorbed energy of gamma or X-ray radiation, in joule per kg matter:
1 Gy = 1 J/kg
The absorbed dose *D* has only to do with the physical effects of radiation, and not with the biological effects, see ‘equivalent dose’ below.

An obsolete unit of the absorbed dose is the *rad*, the acronym of *radiation absorbed dose*, in 1953 redefined as:
1 rad = 100 ergs/g = 0.01 J/kg = 0.01 Gy

Linear Energy Transfer LET

The *Linear Energy Transfer* (LET) is defined as the amount of energy transferred per unit path length by an ionising ray or particle.

$\text{LET} = -\mathrm{d}E/\mathrm{d}t$

The LET is used to quantify the the effects of ionising radiation in organisms and electronic components. In biological and biomedical context the LET is usually expressed in the unit keV/μm (kilo-electronvolt per micrometer). The value of LET varies widely, depending on the speed and electric charge of the involved particle. Due to their relatively low speed and high electric charge (2+) α-particles have generally a much higher LET than β-particles or γ-rays with the same energy.

The biological effects of high-LET radiation are much larger than of low-LET radiation. This because high-LET radiation can transfer more energy with the volume of one living cell, resulting in higher chance of damage to the DNA of the cell.

It is not clear if this model complies with the non-targeted and delayed effects, discussed in section 1.3.

Equivalent dose *H*, unit sievert (Sv)

The biologically effective radiation dose or *equivalent dose H* is defined as the absorbed dose *D* multiplied by the *radiation weighting factor* w_R, in many publications also called the *quality factor Q*, which depends, among other, of the kind of radiation:

$H = w_R \bullet D$

Factor w_R gives the Relative Biological Effectiveness (RBE) of the various kinds of ionising radiation. The biological dose is expressed in the unit *sievert*, symbol Sv. Like the gray the sievert has the dimension Sv [=] J/kg.

1 sievert corresponds with about 1 gray γ-radiation.

Table A1

Weighting factors

radiation	weighting factor w_R
α-rays	20
β-rays	1
γ-rays	1
protons and fast neutrons	10
slow (thermal) neutrons	5

Some other definitions of the equivalent dose are in use as well, which are not discussed here. The sievert Sv is a large unit, for that reason usually the millisievert mSv is used. Generally the assumed lethal dose is 4-5 Sv within a short time.

The old unit of equivalent dose, the biological absorption of radiation, is the *rem*, abbreviation of: *röntgen equivalent in man*. The unit rem is based on the old unit röntgen, which has a definition different from the gray, and is conveniently equated with 0.01 sievert by definition:

1 rem = 0.01 Sv

DARI

The DARI, from the French *Dose Annuelle due aux Radiations Internes,* is the annual radiation dose from internal radioactivity. In human tissue always natural radionuclides are present, with a combined specific activity of about 143 Bq/kg, mainly caused by potassium-40 (^{40}K), 86 Bq/kg, and carbon-14 (^{14}C), 57 Bq/kg, in addition there may be a small contribution of some other radionuclides.
The equivalent radiation dose caused by the natural internal radioactivity is about

$H = 0.17$ mSv/a

of which about 90% is due to potassium-40, about 10% to carbon-14 and a small contribution of other elements
Potassium-40 is a natural radioisotope and a permanent component of living tissue, it emits beta- and gamma rays and has a half-life of 1,3 billion years. One in 8300 potassium atoms is a K-40 nuclide, the other have stable nuclei. Carbon-14 emits only beta rays and has a half-life of 5730 years. It is continuously formed in the atmosphere by interaction of cosmic radiation with atoms of oxygenn, nitrogen high in the atmosphere. Carbon-14 is present in all living organisms

The DARI has been introduced by Charpak & Garwin 2002 [Q300] and defined as:

1 DARI = 0,2 mSv/a

Radiation from natural internal sources is an absolute floor below which it is impossible to sink. In addition man is exposed to radiation from external natural sources: from cosmic radiation, radioactive minerals in rocks and radon-222 seeping from the ground. The contracted doses from natural sources can vary widely, the average is estimated at about 2 mSv per year.

Exposure, unit roentgen (R)

Exposure is a measure of the strength of a radiation field (X-rays or γ-rays) at some point in air. This is the measure made by a survey meter. The commonly used unit of exposure is the roentgen.

The *roentgen*, symbol R, is the old unit of the intensity of ionising radiation from a source, also called *exposure*. The definition dates back to 1928, the year the International X-ray and Radium Protection Committee (IXRPC) was founded, in 1950 restructured and renamed into International Commission of Radiological Protection (ICRP).
The roentgen is the amount of photon energy required to generate $1.610 \bullet 10^{12}$ ion pairs in 1 gram of dry air at 0 °C. A radiation field of one roentgen will deposit acertain amountof electric charge in dry air at standard temperature and pressure (STP). Converted into SI-units the electric charge is measured in coulombs per kilogram dry air and the definition becomes:

$1 \text{ R} = 2.58 \bullet 10^{-4}$ C/kg dry air (STP)

This corresponds with:

1 R = 0.0877 J/kg dry air (STP)

This definition applies exclusively to X-rays and γ-rays and can only be measured in dry air. The reason why the roentgen, not an SI unit, is still widely used stems from the fact that the ionisation of air by ionising radiation is easily measurable with simple equipment (survey meter).

The roentgen is not always appropiate to accurately assess the energy from radiation absorbed by matter. The absorbed energy is an important factor determining the biological damage by radiation, but that amount is not simply measurable.

Summary

Table A2
Units in the radiology

	activity	absorbed dose	equivalent dose (biological effective)	intensity
old units	curie (Ci)	rad	rem	röntgen (R)
SI units	becquerel (Bq)	gray (Gy)	sievert (Sv)	-

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ANNEX **B**

Some important radionuclides

Tritium, carbon-14 and krypton-85, strontium-90, iodine-129

B.1 Introduction

The three radionuclides tritium (^{3}H), carbon-14 (^{14}C) and krypton-85 (^{85}Kr) are routinely released into the human environment by nominally operating nuclear power plants. According to the classical dose-risk paradigm these discharges would have negligible public health effects and so were and still are permitted. This assumption turns out to be untenable based on the evidence of non-targeted and delayed adverse radiation effects, discussed in Chapter 1.

Of special importance are the radionuclides tritium and carbon-14, ^{14}C. As pointed out above these radionuclides are biochemically indistinguishable from their non-radioactive isotopes, normal hydrogen H, respectively normal carbon (mainly ^{12}C). Carbon and hydrogen are two of the six primary building blocks (C, H, O, N, S, P) of proteins and DNA. A complicating factor is that both radionuclides are always discharged simultaneously.

B.2 Tritium ^{3}H

Hydrogen isotopes

Tritium is the radioactive isotope of hydrogen, symbol: ^{3}H, H-3 or T, with a half-life of 12.32 years (values of 12.2 - 12.5 years are also mentioned) and a specific activity of 358 TBq/g. Chemically, tritium is indistinguishable from ordinary hydrogen H, or the other isotope deuterium (^{2}H, H-2 or D). Ordinary hydrogen and deuterium are stable isotopes, of which H is the most abundant: H 99.985% and D 0.015%.

In nature tritium is formed in the upper atmosphere, due to cosmic radiation. Global inventory of natural tritium is estimated at about 2.6 EBq (1 EBq = 1 exabecquerel = 10^{18} desintegrations per second), equivalent with 7.3 kilograms (NCRP-62 1995 [Q251]. The cosmogenic production rate is estimated at about 0.15 EBq/yr.
In other studies global tritium inventories of 2-7 kilograms are reported (Dworschak 1993 [Q80]), of which 1% in the atmosphere and 99% in the oceans as tritiated water HTO. Tritium decays to helium-3 with emission of low-energy beta radiation (electrons):

$$^{3}H \quad \text{—>} \quad ^{3}He + \beta^{-}$$

Anthropogenic tritium production

A hydrogen bomb needs minimal 3 grams of tritium , US warheads contain an average of 5 grams, neutron bombs may contain 10-30 grams (Zerriffi 1996 [Q257]). Due to the decay of tritium, each warhead has to be refurbished after a number of years. Even with a constant number of warheads, tritium production has to be continued.

Thermonuclear explosions in the fifties and sixties of last century added large amounts of tritium to the atmoshpere, peaking at about 115 EBq in 1963. It will decay to about 3 EBq in the year 2030.
The artificial emission of tritium by nuclear installations surpasses the natural generation by cosmic radiation (Dworschak 1993 [Q80]).

Table B1
Some neutron reactions producing tritium or precursors of tritium

^{6}Li	(n,α)	T
^{7}Li	$(n,n\alpha)$	T
^{10}B	$(n,2\alpha)$	T
^{2}H	(n,γ)	T
^{3}He	(n,p)	T
^{9}Be	(n,α)	^{6}Li
^{10}B	(n,α)	^{7}Li
^{12}C	(n,α)	^{9}Be
^{14}N	(n,t)	^{12}C
ternary fission		

Nuclear reactors generate large amounts of tritium. Estimates for LWR's vary from 743-1018 TBq/GWe.a tritium (IAEA-203 1981 [Q74], NEA 1980 [Q75], Dworschak 1993 [Q80], NRC 1996 [Q16]). Most tritium arises from ternary fission in the fuel, the rest from various neutron reactions on light elements present as impurities or components in fuel, coolant and cladding. Chemicals are added to the cooling water, to control the reactivity and the chemistry of the water, in particular boric acid H_3BO_3 and lithiumhydroxide LiOH.

Heavy-water moderated reactors (HWR), like CANDU, produce a hundred times more tritium than a LWR (NCRP-62 1995 [Q251]), see Table 2, mainly due to the D (n,γ) T reaction in the heavy water moderator. It is not clear which fraction of this is annually released at the reactor site. Eventually, all tritium in the moderator will end up in the environment.
Production of tritium in gas-cooled reactors (Magnox, AGR) is about the same as in LWR's (NEA 1980 [Q75]).

Table B2
Calculated production and discharge rates of tritium in LWR's and HWR's in TBq/GW(e).a
Sources: NCRP-62 1995 [Q251], NEA 1980 [Q75]

source	PWR NCRP-62	PWR NEA	BWR NCRP-62	BWR NEA	HWR NCRP-62	HWR NEA
fuel	555-925	750	555-925	750	555	750
coolant	30.7	40	2.3	0	$2.2 \bullet 10^{14}$	$9.0 \bullet 10^{14}$
total		790		750	$2.26 \bullet 10^{14}$	$9.08 \bullet 10^{14}$
discharged *	1.3 + 29.6	4 + 33	0.7 + 1.6	2 + 5	185 + 1850	600 + 150

- Discharges at the reactor site. First number gaseous effluents, second liquid effluents

According to NCRP-62 1995 [Q251], most tritium, about 900 TBq/GWe.a, remains in the spent fuel elements. About 85-90% of the tritium retained the spent fuel elements is discharged into the air and sea at reprocessing plants, when the fuel is dissolved and reprocessed. About 10-15% remains fixed in the Zircalloy cladding, as zirconiumtitride.
If the spent fuel is not reprocessed, this tritium remains in the elements. However, during the long interim storage period of the spent fuel (tens of years), before it is conditioned for final disposal, a part of the remaining tritium slowly diffuses through the Zircalloy cladding and will be discharged as yet.

Chemical properties

Hydrogen atoms and so tritium atoms, are very mobile in the aquatic system and as a consequence in the biochemical system. In tritiated water quickly three chemical equilibria are established:

$$\begin{array}{lll} HTO & < & T^+ + OH^- \\ HTO & < & H^+ + OT^- \\ T_2O & < & T^+ + OT^- \end{array}$$

Exchange reactions with biochemical molecules may occur via various mechanisms, via ions or otherwise, schematically:

$$R_3\text{-}C\text{-}H + HTO \quad < \quad R_3\text{-}C\text{-}T + H_2O$$

When the tritium atom decays to helium-3, a carboniumion may be formed, chemically a very reactive species:

$$R_3\text{-}C\text{-}T \quad < \quad R_3\text{-}C^+ + {}^3He + \beta^-$$

Tritium is discharged into the environment as tritiated water HTO and relatively small amounts of hydrogen gas HT or T_2. Tritium atoms, like ordinary hydrogen atoms, are very mobile in the aquatic system and as a consequence in the biochemical system. They are readily incorporated into biomolecules, which may enter the food chain. Direct ingestion via drinking water, prepared from river water, is another pathway.

Hydrogen atoms bound to the N, O or S atoms in biomolecules of a living cell are easily exchanged with hydrogen atoms from the water molecules in the cell. In a living cell this exchange process goes on continuously. When tritiated water HTO enters a cell, normal hydrogen atoms in the biomolecules are exchanged with tritium atoms. By this process biomolecules with built-in radioactive hydrogen atoms are formed. In this way tritium enters the food chain, for example in milk and vegetables. In the body organically bound tritium (OBT) is slowly formed by metabolic reactions. Once it is formed it stays in the body for much longer periods (20 to 30 times longer) than HTO [24] Fairlie 2008 Q443.

Tritium can enter the body via food as OBT and via drinking water containing tritiated water HTO. The biological half-life of OBT in the human body is much longer than of HTO. Normal hydrogen atoms in the DNA molecules are exchanged for tritium via OBT and HTO. The exposure of DNA to the radiation from tritium comes from within the DNA molecules themselves and from biomolecules and HTO adjecent to the DNA molecules. Despite the short-range of the b-radiation, its effects can be significant. See also [25] NCRP-62 1995 Q251, [26] NCRP-63 1979 Q253, [27] Fairlie 2007 Q373, [28] AGIR 2007 Q444.

If a tritium atom decays during its stay in a DNA molecule it transforms into a stable helium-3 atom, a noble gas, while emitting a beta-particle. The helium atom does not form any chemical bonding and is ejected from the DNA molecule. The emitted beta particle causes additional ionisations and broken bonds in the vicinity of the decay event. Obviously such an event may cause a mutation or lesion in the DNA molecule.

Studies in the 50's, 60's and 70's of the 20th century concluded that deleterious effects of tritiated water from nuclear installations were unlikely at its (then present) levels. This is not to imply that tritium at higher concentrations would be not carcinogenic or mutagenic [26]. Apparently no fundamental, biochemical reaction mechanisms initiated by tritium decay (interaction of bèta radiation with chemical bonds, transmutation and recoil effects) in living organisms have been thouroughly experimentally investigated. Experimental research in mice and rats of biological effects of tritium is reported, but extrapolation to human health effects is difficult. Little direct evidence of tritium effects in humans exists, according to Straume 1991 Q258 [29].

The radiotoxicological classification of tritium is based on theoretical computations, starting from the relatively weak beta radiation of this nuclide [26]. Indications are found that under certain circumstances the biological activity of tritium is higher than predicted [28], [29], [30]. These findings may point to non-targeted and delayed affects.

DNA incorporation

Observations from experiments with mice (1966-1973) suggest that the most hazardous time for mammals exposed to tritium as thymidine, and possibly other DNA precursors as well, would be in utero or as neonatals. No effects following exposure of adult mice were demonstrable in these experiments (NCRP-62 1995 [Q251]).

Health risks

Studies in the 50's, 60's and 70's of the 20th century conclude that deleterious effects of titriated water from nuclear installations appear unlikely at the (then) present levels. This is not to imply that tritium in sufficient amounts would be not carcinogenic or mutagenic (NCRP-63 1979 [Q252]).
Apparently no fundamental, biochemical reaction mechanisms initiated by tritium decay (interaction of bèta radiation with chemical bonds, transmutation and recoil effects) in living organisms are thouroughly experimentally investigated. Experimental research in mice and rats of biological effects of tritium is reported, but extrapolation to human health effects is difficult. Few if any evidence of tritium effects in humans exists (Straume 1991 [Q258]).

The radiotoxicological classification of tritium is based on theoretical computations, starting from the relatively weak beta radiation of this nuclide (NCRP-63 1979 [Q252]). Indications are found that under certain circumstances the biological activity of tritium is higher than predicted (Straume 1991 [Q258], NCRP-89 1987 [Q260]).
The long term effects of a significant higher tritium concentrations in the environment are not clearly understood. On global scale, the rise of the tritium concentrations may seem relatively harmless, but on regional scale relatively high concentrations in surface waters and prolonged exposure may occur. In Western Europe, with numerous nuclear power plants, rivers are used as resources for cooling water for the plants, but the same rivers provide the drinking water for millions of people. We found no empirical data on effects of chronic exposure to tritiated water HTO or tritiated organic material (OBT: organic bound tritium) in humans.

Removal and disposal of tritium

Tritium is released mainly as tritiated water HTO and partly as hydrogen gas HT or T_2. Methods suitable for stripping tritium from large volumes wastes include fractional destillation of HTO-H_2O, cryogenic destillation of HT-H_2 mixes (after electrolysing tritiated water), HT-HTO exchange and H2S-H2O exchange (NCRP-62 1995 [Q251]). After catalytic enrichment the tritium may be immobilized as zirconiumtitride in steel containers (NEA 1980 [Q75]).
Practically, it is not feasible to extract tritium completely from tritriated water. Differences in physical properties of HT and HTO compared to H_2 and H_2O are too small to allow an efficient separation. Moreover, all separation methods are very energy-intensive and require costly equipment.

Theoretically tritiated water can be immobilized in drying agents (e.g. silica gel, polymers, clays) or cement. The solid wastes can packed in appropriate containers and disposed of in a permanent repository. If done, all the cooling water of the reactor were to be immobilized at every occasion the cooling water of the reactor has to be refreshed. Such a procedure would produce large numbers of containers with tritiated waste.
In practice all tritiated water from nuclear power is being discharged into the environment.

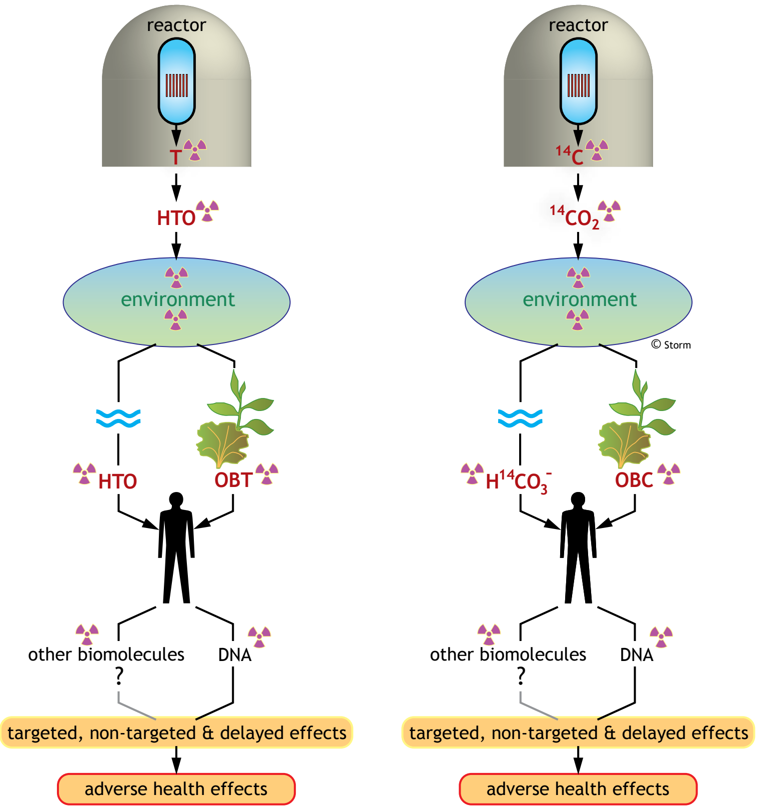

Figure B1
Pathways of radioactive hydrogen (tritium) and carbon-14 into the human metabolism. Both radionuclides are routinely released into the environment by operating nuclear power plants. The pathways are similar. It is generally assumed that damage to DNA molecules cause adverse health effects. Cell damage is not limited to the cells directly hit by radiation, due to the bystander effect (see text).

B.3 Carbon-14

Carbon isotopes

Carbon has two stable isotopes, of which ^{12}C or C-12, ordinary carbon, is the most abundant with98,9%; the other stable isotope is ^{13}C (1.1%). The element has an important radioactive isotope, ^{14}C or C-14, with a half-life of 5730 years (other values also are reported). The specific activity is 0.165 TBq/gram.
In nature ^{14}C is formed in the upper atmosphere by reactions neutrons from cosmic radiation with carbon and nitrogen atoms, mainly by the ^{14}N (n,p) ^{14}C reaction. The global inventory of cosmogenic C-14 in the atmosphere is estimated at 0.14 EBq and the production rate at 0.0014 EBq/yr (NCRP-81 1993 [Q256]). According to NRC 1996 [Q16] the total global inventory is 9.25 EBq, of which about 0.15 EBq in the atmosphere.

Carbon-14 decays with emission of beta rays to nitrogen-14, which is stable:

$$^{14}C \;/\; ^{14}N + \beta^-$$

Anthropogenic carbon-14 production

Table B3
Some neutron reactions producing carbon-14 and precursors

^{13}C	(n,γ)	^{14}C
^{12}C	(n,γ)	^{13}C
^{14}N	(n,p)	^{14}C
^{15}N	(n,d)	^{14}C
^{16}O	(n,γ)	^{17}O
^{16}O	(n,3He)	^{14}C
^{17}O	(n,α)	^{14}C
ternary fission		

Table B4
Calculated production and discharge rates of carbon-14 in LWR's and HWR's, in TBq/GW(e).a
Sources: NCRP-81 1993 [Q256], NEA 1980 [Q75]

source	PWR NCRP-81	PWR NEA	BWR NCRP-81	BWR NEA	HWR NCRP-81	HWR NEA
fuel	0.74	0.4	0.74	0.6		1.0
coolant	0.37-0.52	0.3	0.26	0.4	24	20
core hardware *	1.1-1.6		1.9-2.7			
total	2.2-2.9	4 + 33	2.9-3.7			

* Cladding and structural components of fuel elements

Thermonuclear explosions in the 1950s and 1960s added about 0.36 EBq C-14, so the total global inventory of the atmosphere at present is estimated at 0.50 EBq.

Large amounts of carbon-14 are generated in nuclear reactors, due to neutron-induced reactions of oxygen (in UO_2 and H_2O) and nitrogen and carbon which may be present as components of coolant, moderator, structural materials, fuel, or as impurities.

Calculated production rates in LWR's and HWR's (heavy-water moderated reactors, like CANDU) according to NCRP-81 1993 [Q256] and NEA 1980 [Q75] are summarized in Table 4.
According to NRC 1996 [Q16] spent LWR fuel elements contain about 1.5 Ci ^{14}C per Mg heavy metal (HM), or 0.056 TBq/Mg.

It is assumed that most ^{14}C in cooling water is released at the reactor site and is discharged in the gaseous effluent, less than 1% in the liquid effluent. Very few results of measurements of carbon-14 emissions are published (NEA 1980 [Q75]). In OSPAR 2002 [Q236] not a single measurement of C-14 emission is listed. In NCRP-81 1993 [Q256] one German publication (1982) is mentioned, which reported an annual release of 5.6 Ci/GW(e)y (0.21 TBq/GW(e).y for PWRs and 13.5 for BWRs (0.50 TBq/GW(e).y). The experimental data from which the estimates are made are limited.
If the fuel is reprocessed, the ^{14}C in the fuel will be emitted as $^{14}CO_2$ at the reprocessing plant.

A PWR discharges its carbon-14 mainly as alkanes, over 80% methane and ethane $^{14}CH_4$ and $^{14}C_2H_6$, a BWR in the form of carbon dioxide $^{14}CO_2$ and a few percent of carbon monoxide ^{14}CO and $^{14}CH_4$ (NEA 1980 [Q75]). According to NCRP-81 1993 [Q256], a BWR discharges ^{14}C in a chemical form other than CO_2.
In graphite moderated reactors, e.g. Magnox, AGR and RMBK, much more ^{14}C is produced than in LWR's.

C-14 emission by reprocessing plants is mentioned as problem in WNA inf68 2003 [Q248], but is not mentioned in a more recent version of October 2012.

Chemical properties

Carbon-14 atoms are chemically idendentical with stable carbon atoms (carbon-12 and about !% carbon-13) and enters the biosphere mainly as $^{14}CO_2$. A part is emitted as hydrocarbons (methane, ethane) into the atmosphere, which are slowly oxidized to CO_2. Carbondioxide slightly dissolves in (rain) water and a chemical equilibrium is established:

$$^{14}CO_2 + 2\,H_2O \quad <=> \quad H_3O^+ + H^{14}CO_3^-$$

Via bicarbonate ^{14}C can be bound in the bones of animals and humans.
By photosynthesis in plants, CO_2 is bound into organic materials, which enter the food chain:

$$6\,CO_2 + 6\,H_2O \quad --> \quad C_6H_{12}0_6 + 6\,0_2$$

One or more C atoms may be ^{14}C. In this way also tritium may be built in organic molecules and a combination also is possible. Near a nuclear power plant, both isotopes are present. When a ^{14}C atom decays to a stable nitrogen atom several reactions are possible, e.g.:

$$R_3\text{-}^{14}C\text{-}C\text{-}R_3 \quad --> \quad R_3\text{-}^{14}N^+ + \beta^- + \bullet C\text{-}R_3$$

Both fragments are chemically very reactive.

By the end of the second growing season, most food plants will be nearly at equilibrium with the atmospheric CO_2. The time delay for most foodstuff of plant origin to reach maximum concentration of ^{14}C is not more than one year (NCRP-81 1993 [Q256]).

Soft tissues in animals (and man) seem to lag behind the tropospheric $^{14}C/^{12}C$ ratio by about one to two years. Since some tissues or body compartments have long turnover times, some approaching the life span, total body carbon in the adult subjected to a change in $^{14}C/^{12}C$ intake ratio may not approach equilibrium for decades.

Fetal tissues will more nearly reflect the current ^{14}C composition of the food available to the mother.

DNA incorporation

Male sperm cells are produced from spermatogonia continuously in the adult male and consequently the ^{14}C specific activity in the nuclear material must resemble that of the food with a lag time of one or two years.

Female oocytes are laid down in the fetus before birth and thereafter remain dormant until they ripen just before being shed. It is uncertain how much of the adjacent tissue may be subject to turnover and renewal but, in any case, none of the DNA of the ovum, except the small fraction renewed by repair processes would contain ^{14}C of the current specific activity. There will, therefore, be a delay of 16-40 years in the human female for expression of the genetic effects of a given $^{14}C/^{12}C$ ratio. Calculation of the genetic effect to the current generation of an added increment of ^{14}C to the atmosphere using a lag time of 1-2 years, therefore, is conservative according to NCRP-81 1993 [Q256] .

Suess effect

Before the nuclear age, an equilibrium existed between production and decay of natural ^{14}C, so the $^{14}C/^{12}C$ ratio in the atmosphere (as CO_2) was constant. Via photosynthesis carbon from the air (as CO_2) is integrated in organic material. Generally, every living organism has the same $^{14}C/^{12}C$ ratio in its biochemical molecules. After death, no new ^{14}C atoms are bound in the organism. With time the $^{14}C/^{12}C$ ratio decreases by decay of the ^{14}C atoms. This is the base of the carbon-14 dating method.

Due to the anthropogenic increase of the ^{14}C inventory of the atmosphere, one might expect the $^{14}C/^{12}C$ ratio in living organisms would increase. However, this effect is offset by the increase of the CO_2 content by burning fossil fuels, which are free of ^{14}C. In effect, the $^{14}C/^{12}C$ ratio in the atmosphere decreases by dilution of ^{14}C by stable C; this phenomenon is called the Suess effect, after the investigator who described it for the first time (in 1955).

Carbon has two stable isotopes, of which ^{12}C or C-12, ordinary carbon, is the most abundant (98,9%), the other stable isotope is ^{13}C (1.1%). The element has an important radioactive isotope, ^{14}C or C-14, with a half-life of 5730 years. The specific activity is 0.165 TBq/gram.

Carbon-14 is emitted by a nuclear power plant as $^{14}CO_2$ and $^{14}CH_4$ and some higher alkanes [28]. In air the alkanes are slowly oxidized to $^{14}CO_2$. Carbon-14 atoms are chemically identical with stable carbon atoms and enters the biosphere mainly as $^{14}CO_2$. Carbon dioxide slightly dissolves in (rain) water and a chemical equilibrium is established in which bicarbonate ions are formed. Carbon-14 can be bound in the bones of animals and humans via bicarbonate ions in drinking water.

Via photosynthesis in plants, $^{14}CO_2$ can be bound into biomolecules: the built-in C-14 atoms are called organically bound carbon, or OBC. Via the food chain OBC can enter the human body and become incorporated into DNA molecules. A combination with tritium is possible. Both isotopes are present in the vicinity of all nuclear power plants, because tritium and carbon-14 are discharged into the human environment.

Health risks

By the end of the second growing season, most food plants will be nearly at equilibrium with the atmospheric CO_2. The time delay for most foodstuff of plant origin to reach maximum concentration of ^{14}C is not more than one year [31] NCRP-81 1993 Q256.
Soft tissues in animals (and humans) seem to lag behind the tropospheric $^{14}C/^{12}C$ ratio by about one to two years. Since some tissues or body compartments have long turnover times, approaching the life span, total body carbon in the adult subjected to a change in $^{14}C/^{12}C$ intake ratio may not approach equilibrium for decades.
Fetal tissues will more nearly reflect the current ^{14}C composition of the food available to the mother.

Male sperm cells are produced from spermatogonia continuously in the adult male and consequently the ^{14}C specific activity in the nuclear material must resemble that of the food with a lag time of one or two years.
Female oocytes are laid down in the fetus before birth and thereafter remain dormant until they ripen just before being shed. It is uncertain how much of the adjacent tissue may be subject to turnover and renewal but, in any case, none of the DNA of the ovum, except the small fraction renewed by repair processes would contain ^{14}C of the current specific activity. There will, therefore, be a delay of 16-40 years in the human female for expression of the genetic effects of a given $^{14}C/^{12}C$ ratio. Calculation of the genetic effect to the current generation of an added increment of ^{14}C to the atmosphere using a lag time of 1-2 years, therefore, is conservative according to [31] NCRP-81 1993 Q256.

Damage to DNA molecules occurs in a similar manner as by tritium. A carbon-14 atom decays into a stable nitrogen-14 atom by emitting a beta particle. The chemical properties of nitrogen differ widely from carbon and the beta particle causes secondary ionisations and lesions.
Some DNA molecules in the human body may remain practically unchanged for decades, for example in egg cells, so the medical consequences of carbon-14 ingestion may have long incubation times.

Experiments proved beta-particles (electrons) with remarkably low energy to be able to destroy vital parts of DNA and RNA molecules [32] Collins 2003 Q270. Up until about the year 2000 the common opinion was that DNA and RNA molecules could be damaged only by electrons with an energy of more than 10 eV. Experiments proved that electrons with energies as low as 3 eV can break both strings of the DNA double helix. There is evidence of a lower energy limit well below 1 eV.
The DNA repair mechanism is able to repair a single lesion, but complex and multiple lesions may result in misrepair, ie mutations may occur.

Sparse data are available to base on estimates of genetic hazards associated with ^{14}C-labelled nucleic acid precursors, according to [26] NCRP-63 1979 [Q252] and [30] NCRP-89 1987 [Q260]. This is confirmed by [31] NCRP-81 1993 Q256, which mentiones only a few studies with theoretical calculations, concerning the dispersion of $^{14}CO_2$ in air.

As with tritium, we could not find any evidence that fundamental, biochemical reaction mechanisms initiated by carbon-14 decay (bèta radiation, transmutation and recoil effects) in living organisms are thouroughly experimentally investigated.

The radiotoxicological classification of carbon-14 is based on theoretical computations, starting from the relatively weak beta radiation of the nuclide, and not on empirical biological evidence.

Long term effects of a significant higher carbon-14 concentration in the environment are unknown. On global scale, the rise of the concentrations may seem insignificant, but on regional scale relatively high concentrations in air, water and plants may occur.

Removal and disposal

Removal of ^{14}C from gaseous effluents requires catalytic oxidation of hydrocarbons and CO to CO_2. The CO_2 may be removed from the off-gas with a solvent-absorption process. The CO_2 so collected could be discharged into a slurry of calciumhydroxide $Ca(OH)_2$ and converted to calciumcarbonate $CaCO_3$.
Another technique is the double caustic scrubbing process: absorption of in sodium hydroxide solution and then reaction with calcium hydroxide solution for precipitating the carbonate from the first solution as calciumcarbonate (NEA 1980 [Q75]).
The $CaCO_3$ can be immobilized using several agents, e.g. cement, for permanent disposal.

B.4 Krypton-85

Krypton is one of the six inert gases (noble gases), the other five being helium, neon, argon, xenon and radon. The atmosperic abundance is 1.14 ppm (volume).
There are six stable isotopes of krypton and a number of radioactive isotopes, of which krypton-85 (^{85}Kr or Kr-85) is the most important.
The half-life of ^{85}Kr is 10.7 years, the specific activity 14.5 TBq/gram. It decays with emission of beta and gamma rays to stable rubidium-85.

$$^{85}\mathrm{Kr} \longrightarrow {}^{85}\mathrm{Rb} + \beta^- + \gamma$$

Krypton-85 is present in the natural environment due to spontaneous and neutron-induced fissions of natural uranium and neutron capture by stable ^{84}Kr of cosmogenic neutrons in the upper atmosphere. Calculated world equilibrium inventory is about 0.08 TBq in the upper three meters of total land and water surface and about 0.4 TBq in the atmosphere.

Anthropogenic krypton-85 production

Nuclear weapons test (1945-1962) added about 0.19 EBq (1.9×10^5 TBq). The current world inventory of ^{85}Kr is calculated at about 0.074 EBq (NCRP-44 1975 [Q259]).
In nuclear reactors large amounts of krypton-85 are produced, as one of the major fission products.

Table B5
Calculated production and discharge rates of Kr-85 in LWR's and HWR's, in TBq/GW(e).a
Sources: NCRP-44 1975 [Q259], NEA 1980 [Q75]

source	PWR NCRP-44	PWR NEA	BWR NCRP-44	BWR NEA	HWR NCRP-44	HWR NEA
fuel	11000	14000	11000	14000	11000	14000
coolant	0	0	0	0	0	0
discharged *	<110	10	<110	10	<110	10

- Discharges at the reactor site in gaseous effluents

Chemical properties

Krypton, like the other noble gases, is tasteless, colorless and chemically (almost) inert. A few fluor compounds of krypton are synthesized at cryogenic temperatures. The gas has a low solubility in water and a high solubility in nonpolar solvents. Noble gases can be bound in clathrates in which the atoms are physically entrapped in cages of organic molecules.
The decay product rubidium is an alkali metal and has similar chemical properties as potassium.

Biological properties

Being chemically inert, krypton and the other noble gases are not usually involved in biological processes. They are, however, absorbed into the tissues of the body via inhalation and dissolution in body fluids and tissues. Xenon has been shown to combine with specific sites with certain

protein molecules. Krypton is characterized by low blood solubility, high lipid solubility and rapid diffusion in tissue.
Exceptions to the the biologically inert characterization of inert gases have been noted by numerous studies. A comparatively high uptake of krypton by the adrenal gland has been reported. These phenomena are not understood (NCRP-44 1975 [Q259]).

Health risks

On global scale the genetic and overall carcinogenic effects from Kr-85 are calculated to be small as compared with other possible sources of deleterious effects.
The possible interaction of radiation from krypton-85 and solar ultraviolet (UV) should be mentioned. In order to understand better the implications of long-term ^{85}Kr releases to the atmosphere, epidemiological and laboratory studies should be undertaken to define the nature and degree of interaction, if any, of UV radiation with ionizing radiation in the induction of skin cancer (NCRP-44 1975 [Q259]).

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Removal and disposal

Absorption on charcoal or moleculair sieves at ambient temperatures is only suitable for delaying the release of waste gases and not effective for control of ^{85}Kr.
Absorption on charcoal at liquid nitrogen temperatures is adaptable for recovery of krypton. Because the beds are cooled and heated alternatively, energy consumption for refrigeration is high. The system has several other severe disadvantages.
Cryogenic destillation is to be one of the more promising processes for krypton and xenon recovery. The gases can be stored in pressurized cylinders. This process also has a relatively high specific energy consumption.
Selective absorption or extraction by fluorocarbon solvents has been tested.
On a laboratory scale other processes have been investigated: recovery by permselective membranes and clathrate precipitation from organic solvents (NCRP-44 1975 [Q259], NEA 1980 [Q75]).

In practice all krypton-85 is released into the atmosphere.

B.5 Strontium-90

nn

B.6 Technetium-99

One of the troublesome radionuclides is technetium-99, The most stable form of this nuclide in water is the pertechnetate ion $^{99}TcO_4^-$ ion. In the human body this ion accumulates in the thyroid gland as it mimics the iodide ion I^-.

^{99m}Tc $T^1/_2$ = 6.0 hours

^{99}Tc $T^1/_2$ = 2.13×10^5 years fission yield 628 g/Mg fuel 25000 MWday/Mg => present fuel about double burnup and double yield => about 1.2 kg/Mg fuel

Pertechnetate compounds are highly soluble in water and it is difficult to extract pertechnetate ions from waste liquids. Removal of fission products is based on cation reactions, such as $^{137}Cs^+$, so an additional anion-based separation step is required.

B.7 Ruthenium-106

nn

Removal of fission products is based on cation reactions, such as $^{137}Cs^+$, so an additional anion-based separation step is required.

B-8 Iodine-129

nn

B-9 Cesium-137

nn

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ANNEX C

Uranium

C.1 Uranium mining

Uranium is a radioactive metal, which decays by alpha and gamma emission into other elements, called the decay products or ‘daughters’. The decay products are also radioactive, most of which are potent alpha-emitters. The final decay product is stable lead. Consequently uranium bearing rock contains a number of radioactive elements. In the natural condition the radionuclides are confined in more or less insoluble minerals in the rock of the uranium ore deposit. Uranium ores are generally very old geologic formations, with ages of billions of years.

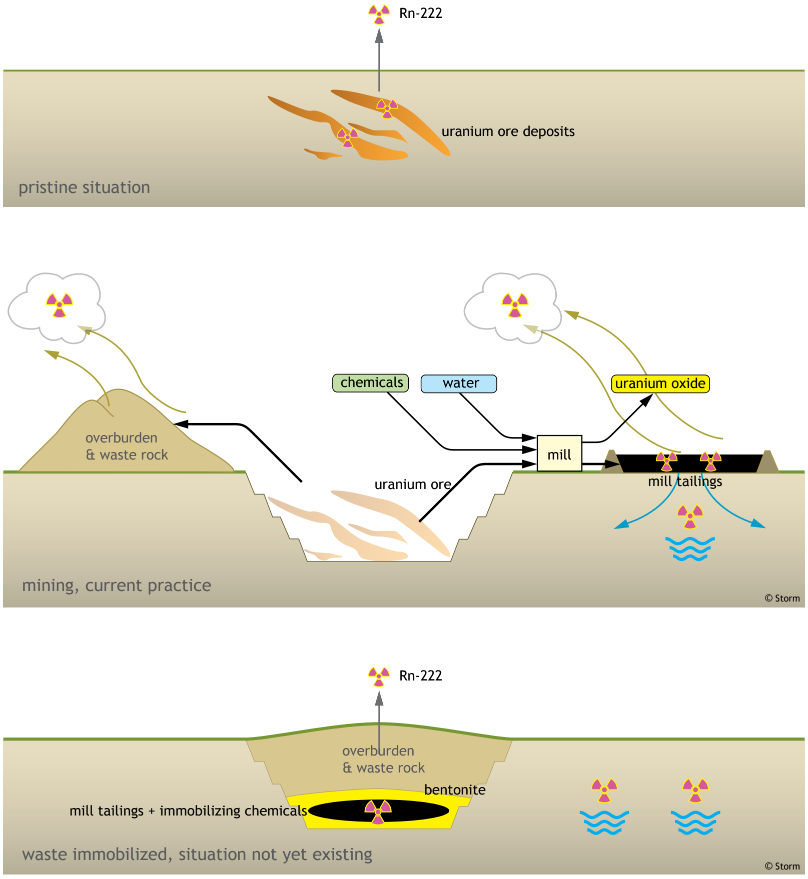


Figure C1
Outline of uranium mining, the first step of the nuclear process chain. The area directly disturbed by the mining operations of a large uranium mine may come to some 100 km^2. The indirectly disturbed area, by wind blown dust and contaminated groundwater, may run into hundreds of thousands of square kilometers. When the ore deposit is exhausted, the dangerous mill tailings should be immobilized and the mine and its surrounding area should be restored to the original situation, a process called mine reclamation, see text. The ground water table remains contaminated permanently.

In spite of its old age uranium-bearing rock is anything but harmless, for the rock emits gamma radiation and the dust of it contains dangerous alpha emitters. One of the decay products is the radioactive noble gas radon-222 that escapes into the air. When groundwater enters an ore deposit, the radioactive minerals may slowly disintegrate and as a result the groundwater will be contaminated with radioactive species. By far the most uranium deposits of the world are located in sparsely inhabitated and often arid areas, for example in Australia, Namibia, Kazakhstan and USA.

To obtain uranium it has be extracted from uranium ore by physical and chemical separation processes. At the uranium mine the ore is mined, then milled (ground to powder) and finally chemically treated to extract the uranium. The other radionuclides in the ore, the decay products of uranium, remain in the tailings (waste stream) of the extraction process. The mill tailings have the appearance of a watery mud and consist of the ore powder, chemicals and large volumes of water. The radioactive mud is stored in large ponds (see Figure C2).
A part of the water from the mud will evaporate and the other part, including the dissolved radionuclides, drains into the ground. When the mill tailings go dry, the remaining fine powder will be easily spread by the wind. This situation occurs when one pond is filled up and a new one is taken into operation and after the mine has been mined out and is abandoned.
Satellite photo's show dust from the Sahara desert crossing the Atlantic Ocean under certain conditions. An indication how far dust, and so radioactive dust, can be transported by the wind.

Apart from the remaining uranium isotopes (U-238, U-235, U-234) in the mill tailings – the extraction yield or recovery always is less than 100% – the mill tailings contain the uranium decay daughters. Some uranium ores contain a significant fraction of Th-232 and its decay products. The activity of some nuclides in mill tailings are given in Table C1, taken from [37] INFCE-7 Q277 and [38] Diehl 2006 Q343.

Table C1

Activity of the radionuclides of the U-238 decay series in mill tailings (in terabecquerel TBq), set free per GWe.a electricity production by LWR nuclear power plants. The second column gives figures from INFCE-7 1980 Q277 [37]. The third column gives figures in Bq/g from Diehl 2006 Q343 [38], valid for ore at a grade of 0,1% U and an extraction yield of 90%. In the fourth column the figures of Diehl are converted into TBq/GWe.a, to make them comparable with the figures of INFCE-7. Assuming the reference reactor consumes 200 Mg (metric tonnes) of natural uranium per GWe.a, the mass of the mill tailings is 200000 Mg/GWe.a at an ore grade of *G* = 0.1% U. The world average grade of mined uranium ore in 2012 was 0.1-0.05% U.

nuclide	activity TBq/GWe.a (INFCE-7)	activity Bq/g (Diehl)	activity TBq/GWe.a [34], [1]	decay mode
U-238	1.3	1.2	0.24	alpha-emitter
Th-234	-	1.2	0.24	beta-emiiter
Pa-234m	-	1.2	0.24	beta-emiiter
U-234	1.3	1.2	0.24	alpha-emitter
Th-230	25.2	12.3	2.5	alpha-emitter
Ra-226	26.6	12.3	2.5	alpha-emitter
Rn-222	-	12.3	2.5	alpha-emitter
Po-218	-	12.3	2.5	alpha-emitter
Pb-214	-	12.3	2.5	beta-emiiter
Bi-214	-	12.3	2.5	beta-emiiter
Po-214	-	12.3	2.5	alpha-emitter
Pb-210	26.6	12.3	2.5	beta-emiiter
Bi-210	-	12.3	2.5	beta-emiiter
Po-210	-	12.3	2.5	alpha-emitter

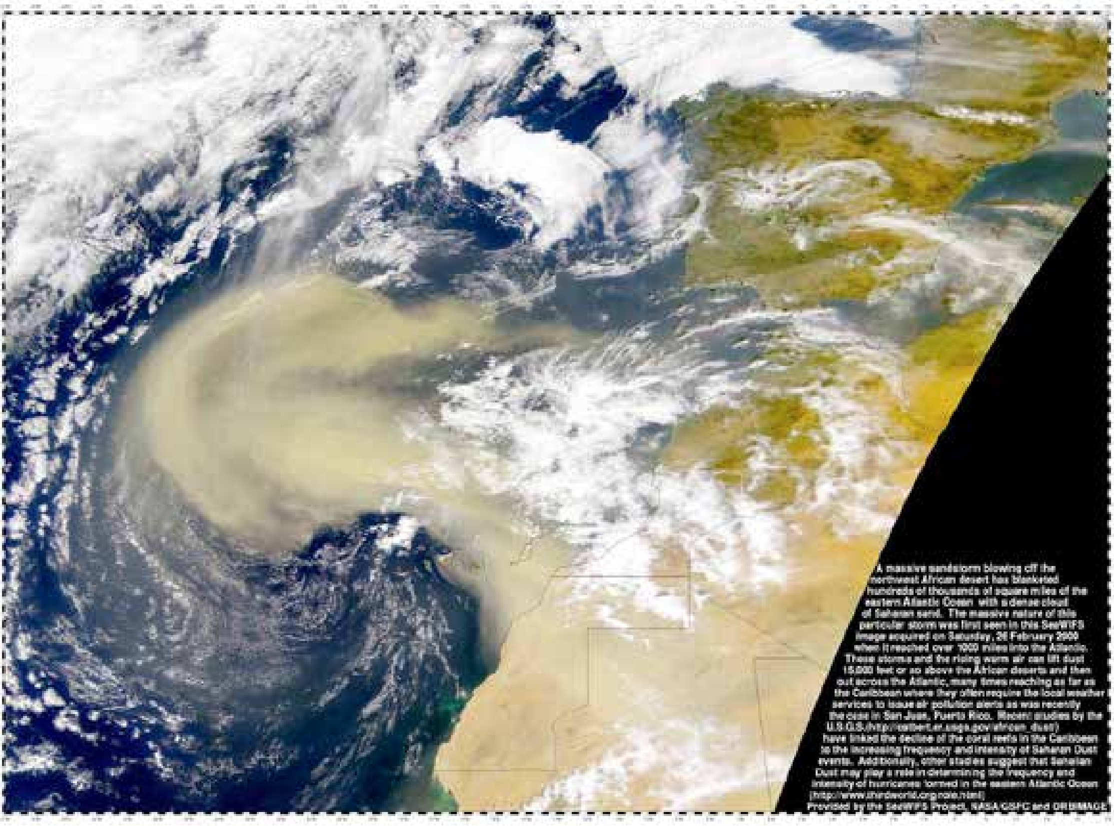
INFCE-7 [37] does not specify the ore, extraction yield and natural uranium consumption per GWe.a the table is based on. It is not clear why the activities of other radionuclides are not given by INFCE-7, neither why their figures are higher than those of Diehl [38] by a factor ten.

Figure C2
Satellite photo of the Ranger uranium mine in Australia, a medium-sized and one of the cheapest operating uranium mines in the world. The large green geometric pond on the lower left is the mill tailings pond. The light colored areas are the overburden and waste rock dumps. The active mining pit is the one at the upper right, partially flooded. The round dark object at the lower center is a former, mined out, pit which is fully flooded. The extraction plant (mill) is on the far right, barely discernable. Note the scale bar in the lower left corner. Source photo: Google Maps.

The dangerous decay products of the uranium isotopes are physically and chemically mobilized. The dust blown off the mill tailings contains highly radiotoxic elements, such as Ra-226, Pb-210 and Po-210. The lethal dose of polonium-210 is some 50 nanogram. Inhalation of the dust is a dangerous contamination pathway, for most of the radionuclides are potent alpha emitters. The decay products can also enter the body via drinking water, as the groundwater at large distances from the mining area may be contaminated with soluble compounds of the radionuclides, seeping from the mill tailings. By the way, the escaping radon-222, a noble gas, from uranium bearing rock offers one of the methods to localize uranium ore bodies.

All radionuclides present in the tailings pose a serious long term environmental risk ([16] NRC 1996 Q16, [39] Andriesse 1994 Q77, [40] Lipschutz 1980 Q54), all the more so because the elements are chemically mobile after the milling process. The publication of Blanchard et al. 1982 [41] deepens that worries. Health risks from uranium mining are also addressed by Diehl 2006 Q343 [38]. Despite the alarming studies from as early as the 1980s, the nuclear industry still does not consider uranium mining to be a health risk for many millions of peaople.

A study from 2006 (http:///www4.nau.edu/insidenau/bumps/2006/2_22_6/uranium.htm) [42] found that uranium can also damage DNA as a heavy metal, independent of its radioactive properties. When cells are exposed to uranium, the uranium binds to DNA and the cells acquire mutations.



A massive sandstorm blowing off the northwest African desert has blanketed hundreds of thousands of square miles of the eastern Atlantic Ocean with a dense cloud of Saharan sand. The massive nature of this particular storm was first seen in this SeaWiFS image acquired on Saturday, 26 February 2000 when it reached over 1000 miles into the Atlantic. These storms and the rising warm air can lift dust 15,000 feet or so above the African deserts and then out across the Atlantic, many times reaching as far as the Caribbean where they often require the local weather services to issue air pollution alerts as was recently the case in San Juan, Puerto Rico. Recent studies by the U.S.G.S.(http://catbert.er.usgs.gov/african_dust/) have linked the decline of the coral reefs in the Caribbean to the increasing frequency and intensity of Saharan Dust events. Additionally, other studies suggest that Saharan Dust may play a role in determining the frequency and intensity of hurricanes formed in the eastern Atlantic Ocean (http://www.thirdworld.org/role.html)
Provided by the SeaWiFS Project, NASA/GSFC and ORBIMAGE

Figure C3
Satellite photo of a massive sandstorm blowing off the northwest African desert. Source: SeaWIFS Project, NASA GSFC and ORBIMAGE.

C.2 Mine reclamation

Strikingly the dangers from uranium mining get little attention in the media. One reason may be the fact that by far the most uranium mines are located in sparsely inhabitated areas, most of which have an arid climate. So it can happen that people in Sydney and Melbourne in Australia inhalate radioactive dust from the Olympic Dam mine, the largest uranium mine in the world.
Mine reclamation is never included in the cost pictures of nuclear energy, nor in the published energy balances of the nuclear chain, except in [1] Storm & Smith 2008 Q6. The authors of the Australian study ISA 2006 Q325 [43] considered mine reclamation, although a hot item in Australia, to be outside the scope of their study.

Mine reclamation comprises the actions needed to restore the mining area to a habitable one

again. The chemically mobile radionuclides in the mill tailings should be immobilized again and put back in the mining pit, as deep as possible. To prevent remobilization by groundwater flows, the mill tailings must be shielded from the groundwater by an effective barrier.
The authors of [1] Storm & Smith 2008 Q6 proposed the following concept to limit the health effects of uranium mining:
The mill tailings are mixed with immobilizing chemicals, for example sodiumphosphate, for most phosphates are highly insoluble in water, and the resulting mass is put back into the mining pit between thick layers of bentonite (see also Figure C1). Bentonite is a clay mineral with special properties: it swells by the uptake of water, effectively closing fissures and microchannels, and has strong ion-exchange properties, resulting in a very low migration rate of nuclides other than hydrogen ions and some alkali metal ions.
In view of the exceedingly large masses of the mill tailings (often tens of millions of tonnes at each uranium mine), above concept seems reasonable. As said before, it is a hypothetical concept, for it has never been tried in practice. To our knowledge no other study ever included a concept of mine reclamation.

Nowhere in the world, as far as known, the impact on the environment by uranium mining has been compensated for in a way that can be considered ecologically adequate and safe to the local inhabitants. Uranium mining companies leave the mill tailings unshielded in the mining area. After the last kilogram of uranium has been removed from the site, the lights are turned off and the gate is closed.

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ANNEX **D**

Reprocessing and nuclear safety

D.1 Vital role of reprocessing in advanced nuclear concepts

Spent nuclear fuel from a light-water reactor (LWR) contains a large fraction of uranium-238, a part of the original uranium-235 remaining unfissioned, fission products, plutonium and trans-plutonium actinides. Both plutonium and the higher actinides originate from uranium by neutron capture. Spent fuel is an exceedingly complex mixture of nuclides, representing almost the complete Periodic System of the Elements, and is highly radioactive. The Zircalloy cladding of the fuel elements has also become highly radioactive, by neutron capture.

Separation of spent fuel into fractions is possible by an intricate complex of chemical processes, called reprocessing. Reprocessing is the pivotal process in several nuclear concepts:

1 plutonium for weapons
2 breeder reactors (U-238/Pu-239 cycle)
3 thorium reactors (in fact the Th-232/U-233 breeder cycle)
4 plutonium recycling in LWRs
5 radioactive waste volume reduction by vitrification
6 partitioning & transmutation, to convert long-lived radionuclides into short-lived.

Actually concepts 4 and 5 could be combined, as well as concept 2 with concept 6.

Initially reprocessing has been developed in the early days of the nuclear age to produce plutonium for atomic weapons. In later years commercial applications of the reprocessing technology have been developed from the military applications, when de breeder concept came into the picture. Main purpose of the civil reprocessing plants, in Europe at La Hague in France and Sellafield in Great Brittain, was to get the plutonium, to fuel fast breeder reactors (FBR's) and to recycle unused uranium.

Before addressing concepts 2-6 (military applications of nuclear technology are not discussed here), first follows a introduction of the civil reprocessing technology. In this introduction some topics are briefly discussed: investments of energy and materials, monetary cost, inherent limitations of reprocessing, authorized radioactive discharges into the environment from reprocessing plants and the risk of accidents involving very large amounts of radioactivity.

D.2 Reprocessing of spent nuclear fuel

The process

Reprocessing of spent fuel is an exceedingly intricate complex of physical and chemical separation processes. In principle two main routes for reprocessing/partitioning are possible: the hydrochemical way, which is based on chemical processes in aquous solutions, and the pyrochemical way, based on processes in molten salts (mainly fluorides) at high temperatures. The molten-salt method is proposed for combination with advanced concepts of molten-salt reactors in an integrated system. There have been done some experiments with molten-salt reprocessing, but its operational application is very remote. All existing reprocessing plants are operating with water-based separation processes.

In the first phase of the sequence the spent fuel is chopped into small pieces and treated with boiling nitric acid. The nuclear fuel dissolves into the liquid, the cladding hulls do not. In addition some insoluble compounds from the fuel remain in solid state. The insoluble fraction is separated from the solution; this fraction becomes larger as the burnup of the fuel is higher. Burnup is a measure of how much energy is extracted from a given amount of nuclear fuel.

In the second phase of the reprocessing sequence uranium and plutonium is separated from the solution resulting from phase 1, together with other actinides.

In the third phase the recovered uranium and plutonium are separated from the other actinides and purified, by means of various extraction processes.

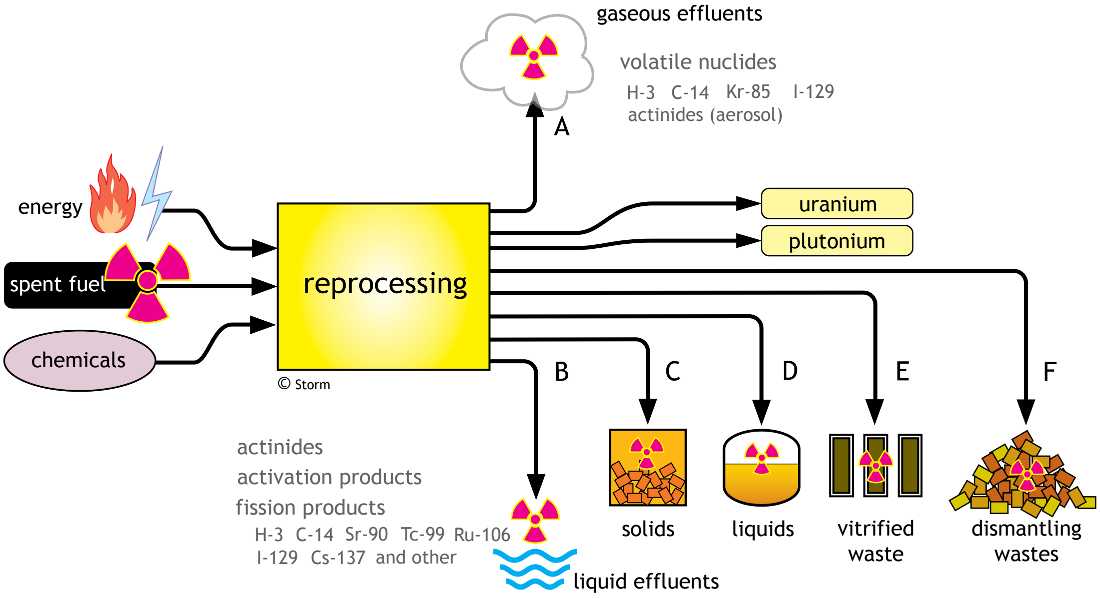

Figure D1

Outline of the radioactive mass flows of reprocessing. The input of a reprocessing plant consists of spent fuel, chemicals and energy (electricity and fossil fuels). Spent fuel is separated into seven fractions: unfissioned uranium, newly formed plutonium and five waste fractions A-E:

A gaseous effluents, discharged into the atmosphere, containing gaseous and volatile fission products, activation products, noble gases and some aerosols of other fission products and actinides,

B liquid efflents, discharged into the sea, containing some U and Pu and other actinides, in addition to a substantial part of the highly soluble fission products; this fraction is to be vitrified,

C insoluble solid waste consisting of spent fuel cladding hulls and other solids, containing small amounts of U, Pu, fission products, activation products and actinides,

D liquid wastes containing fission products, activation products, uranium, plutonium and other actinides, resulting from imperfect separation and purification processes.

E the fraction of fission products, activation products and actinides which can be vitrified.

An eighth radioactive waste stream, fraction F, consisting of dismantling wastes, will be released after final shutdown of the reprocessing plant, when the plant will be decommissioned, cleaned and dismantled.

Reprocessing and the Second Law

Reprocessing of spent fuel is a sequence of separation processes, involving numerous chemical equilibria and complicated by high radiation levels. Nuclear radiation causes radiolysis of the solvents and extraction liquids, which results in less effective separation and the generation of unwanted chemical species.

Separation processes are governed by the basic laws of nature. One of the consequences of these laws is that separation processes never go to completion. For that reason it is impossible to separate a mixture of *n* different chemical species into *n* 100% pure fractions without losses. Separation becomes more difficult and goes less completely as:

- more different kinds of species are present in the mixture,
- the concentration of the desired species in the mixture are lower,
- constituting species are chemically more alike
- the solution is higher radioactive.

Complete separation is a fiction. As a consequence of these features a part of each desired fraction will be lost in the waste streams and each fraction will be contaminated with species from other fractions. The selectivity of separating a certain fraction from a mixture can be enhanced, at the expense of more specialistic chemicals and equipment and consequently more energy, and more losses of other fractions.

Radioactive and non-radioactive isotopes of the same element cannot be separated.

The amount of radioactivity in spent fuel does not change by the mechanical and chemical treatments in the reprocessing plant, it simply means a reshuffling of the radionuclides from one material flow to several other. Inevitably, mixing an amount of radionuclides, compacted in a solid (spent fuel), with nonradioactive fluids or other substances increases the volume of the radioactive waste, complicating the waste disposal problems.

As pointed out above separation of the elements in a solid or solution never can be complete, partly due to the chemical properties of the components of a mixture, partly due to inherent chemical and physical aspects of extraction equilibria, partly due to technical imperfections. Economic considerations and the human factor are left aside here. The difficulties increase with the number of compounds or elements in the mixture which are to be separated.

A single substance can be made very pure, but at the expense of

- inevitable loss of a portion of that substance into the waste streams,
- consumption of large amounts of chemicals,
- consumption of large amounts of energy,
- the need for complex equipment.

Costs

The costs of reprocessing are high and escalating rapidly. Large cost escalations are norm for all advanced technologies [RAND 1979] [Q127] and [RAND 1981] [Q126]. [NRC 1996] [Q16] estimated the costs at 2110 $(1992)/kg HM (about 2600 $(2000)/kg HM). HM stands for heavy metal: uranium + plutonium. The cost of plant decommissioning has been neglected in this estimate.

Storm 1985 Q2 published a graph with the historical cost of reprocessing with entries from more than 20 publications. In 1983 the rapidly rising costs (escalation 50-60% per year) approached 10000 $(1982)/kg HM (about 18000 $(2000)/kg HM), including the construction of the reprocessing plant, but excluding dismantling of the facilities.

For the reprocessing plant at Sellafield (UK) the preliminarily cost estimates vary from GBP38bn (€45bn) (NDA 2009 [Q501]) [99]n to GBP50-100bn (€60-120bn) [82] (*Nature*, 23 November 2006 p 245) [100]n and will take some 130 years. Assuming the decommissioning and dismantling of the Sellafield reprocessing plant will cost €100bn and its lifetime spent fuel throughput was 10000 Mg (metric tonne), the contribution to the reprocessing cost from decommissioning and dismantling alone would be 10000 €(2006)/kg HM. Most likely in practice these cost will rise to a multitude of this value: cost overruns are the rule in the nuclear industry (see also section 9.2).

The decommissioning and dismantling of the US West Valley reprocessing plant, which operated from 1966-1972 and reprocessed 640 tonnes of spent fuel, will cost from 2007 on at least €4bn (€(2007)) and will take another 40 years to complete. Very likely the final cost will be considerably higher. Up until 2007 several billions of dollars already have been spent on West Valley (UCS 2007 [Q421]) [49]n. Above figures point to a specific dismantling cost of some 10000 €(2007) per kilogram reprocessed heavy metal (HM).

How viable is reprocessing from an economic point of view?
We found no indications that LWR spent fuel will be reprocessed in the future on a significant scale, once the existing contracts expire; see also MIT 2003 [Q280].

D.3 Discharges from reprocessing plants

A part of the fission products is gaseous (e.g. noble gases) or easily form gaseous compounds and escape from the spent fuel solution. Most of those nuclides are difficult, and consequently costly, to retain chemically into solid materials or physically in containers, notably tritium (mainly as HTO), carbon-14 (mainly as $^{14}CO_2$), iodine-129 (various compounds, e.g. as $H^{129}I$ or $^{129}I_2$) and the noble gases such as krypton-85 and xenon-133. The nuclides are discharged into the air or sea. In addition a significant fracion of the highly soluble radionuclides are discharged with the liquid effluents, notably Sr-90, Tc-99, Ru-106 and Cs-137.

Table D1
Discharges of radionuclides in the liquid and gaseous effluents of a reprocessing plant. Sources: NEA 1980 [Q75], Pigford et al. 1973 [Q112] and various NCRP reports.

nuclide	NEA 1980 TBq/GWe.a	Pigford 1973 TBq/GWe.a	NCRP TBq/GWe.a	NCRP reference
H-3	630	885	555-925	NCRP-62 1995
C-14	0.4 - 0.6	-	0.74	NCRP-81 1993
Kr-85	14000	13800	11000	NCRP-44 1975
Ru-106	-	0.136	-	
I-129	0.05	0.022	0.042	NCRP-75 1983
other fission products	-	0.340	-	
transuranics	-	0.00014	-	

Table D2
Discharge limits of radionuclides from reprocessing plants into the sea in 2000. Source: OSPAR 2002 Q236.
n.a. = not available

radionuclide	discharge limits La Hague TBq/yr	discharge limits Sellafield TBq/yr
tritium H-3	37000	25000
total alpha	17	1
total beta	1700	400
plutonium	n.a.	0.70
uranium	n.a.	2000 kg/yr

The authorized discharges limits seem to be dimensioned in such way, that at nominal operating capacity the reprocessing plants are allowed to discharge all hardly (= costly) retainable radionuclides into the environment.
Not by chance the European reprocessing plants (La Hague in France and Sellafield in the UK) are located at the sea coast.

Table D3

Discharge limits of radionuclides of the reprocessing plant at La Hague. Source: [51] Malherbe 1991 [Q17] [36]n. n.a. = not available

radionuclide	gaseous effluents TBq/yr	liquid effluents TBq/yr
tritium H-3	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

Published discharges lower than the permitted limits do not necessarily mean a better retainment of the radionuclides, but might more likely point to a lower throughput of the plant, or to a lower burnup of the reprocessed spent fuel, containing less radionuclides per kg.

D.4 Plutonium recycling

Closed-cycle reactors

What is called a ‘fast breeder reactor (FBR)’, or ‘fast reactor with closed fuel cycle’ is not just a reactor type or a stand-alone system. To exploit fully the promised potential of fissioning some 60% of the nuclei in natural uranium, a complex breeder cycle is a prerequisite, consisting of three components: breeder reactor, reprocessing facility and fuel fabrication plant. Each of the three components has to operate flawlessly according to the design specifications and, moreover, finely tuned to each other, in order to let the system actually breed more fissile nuclei from non-fissile uranium-238 than it consumes. If one component of the cycle operates less than flawlessly, the whole system fails and the breeding gain gets lost.

Fifty years of intensive research in a number countries, e.g. USA, UK, France, Germany, former USSR now Russia, Japan and India, with investments of hundreds of billions of dollars so far have failed to demonstrate that the breeder (U-238/Pu-239) cycle is technically feasible. No technical breakthroughs are reported contradicting the observation of the unfeasibility of the breeder cycle. This is no surprise, because the main problems preventing the realization of a closed-cycle reactor functioning according to the promises are of fundamental sort. The concepts of closed-cycle reactors, the ^{238}U-^{239}Pu breeder as well as the ^{232}Th-^{233}U breeder, are implicitely based on the assumption that perfect materials, perfect equipment and perfect separation processes could become available. Exactly these conditions are impossible, as a consequence of the Second Law of thermodynamics, as will be explained in Annex F. The closed-cycle fast reactor is discussed in more detail in Annex E.

Since the FBR proved to be unfeasible, spent fuel reprocessing essentially became superfluous. The only raison d'être of reprocessing would be recycling of uranium and plutonium in LWR's, but that is questionable either (MIT 2003 [Q280]).

Plutonium in LWRs, MOX fuel

Plutonium can be 'burned' in LWR's, using MOX fuel (Mixed OXide). In that case the uranium is 'enriched' with Pu, instead of U-235. Most reactors using MOX fuel have about 30% of the core fuelled with MOX elements. More than 50% MOX requires a special design of the reactor (WNA inf29 2003 [Q246]).

During reactor operation, fissile plutonium nuclei (Pu-239 + Pu-241) in the MOX fission. Simultaneously new plutonium nuclei arise from uranium-238 nuclei by neutron capture. By the same process higher actinides are formed from plutonium. The net result depends on the proportion of MOX fuel in the reactor core. In the present situation, hardly more than a steady state may be reached with Pu recycle: the system then burns as much plutonium as it generates. MOX fuel cannot be reprocessed (WNA inf13 2003 [Q245]), or only once or twice (WNA inf15 2002 [Q247]), because of deteriorating isotopic quality of the plutonium. Nonetheless, recycling of LWR MOX fuel is considered as means of destruction of plutonium and as energy source. A MOX cycle lasts at least 11 years, which may be a problem in itself (NEA ppr 2003 [Q249]).
The production MOX fuel is costly, because of the high radioactivity of de components. MOX fuel complicates power plant operation for utilities, introducing more risks and higher costs.

No net energy from MOX

From an energy point of view, reprocessing of spent fuel and using MOX fuel means introducing two processes into the nuclear chain, both of which are very energy-intensive. MOX fuel itself can't be recycled, because of the degraded isotopic composition of both the reprocessed uranium and the reprocessed plutonium.
No net energy gain can be expected by the use of plutonium in LWRs, because the introduction of the two additional processes – reprocessing and MOX fuel fabrication – into the nuclear fuel chain are very energy-intensive. The energy balance becomes strongly negative if the decommissioning and dismantling energy investments are also accounted for.

Health risks from MOX fuel

The use of MOX fuel in commercial nuclear power plants poses serious health and safety risks, as pointed out in section 6.3.

D.5 Vitrification of radioactive waste

Waste management concepts

In its communication with politicians and the general public the nuclear industry advocates several options to solve the radioactivity immobilisation problem. None of the proposed solutions have become operational up until this moment, 70 years after the first human-made radioactive waste had been generated.
One of these options is the disposal of spent fuel elements, packed in durable containers, in a

geologic repository and is discussed in Chapter 2. Two other proposed solutions are based on sophisticated technical concepts to deal with the human-made radioactivity:

- Radioactive waste volume reduction by vitrification, aimed at reducing the volume of high-level radioactive waste. This concept will be discussed in this section.
- Partitioning & transmutation, claimed to be able to reduce the amount of high-level waste. This concept will be addressed in Annex E.

According to a popular view within the nuclear industry both technical concepts could reduce the high-level waste problem to a routine job, nothing to worry about, see for example MacKay 2009 Q399 [15].

Both concepts may seem plausible at a first glance. However, on closer examination their feasibility as radioactive waste reduction strategies proves to be based on fallacies, ignoring one of the most basic laws of nature: the Second Law of thermodynamics (see Annex F).

Vitrification

Vitrification is advocated as a means of a significant volume reduction of radioactive materials removed from a nuclear reactor. According to this concept long-lived fission products and the minor actinides, the radionuclides from spent fuel considered to be the most dangerous, would be chemically immobilized in a matrix of borosilicate glass. The fission products and actinides (other than U an Pu) then are converted into oxides, which are mixed with a glassmaking frit and melted to form a borosiicate glass. The glass is poured into appropiate stainless steel containers, which are to be placed in a geological repository for permanent disposal. In other processes, e.g. Synroc, the wastes are mixed wich several metaloxides and converted into a crystalline ceramic material.

Some nuclides are discharged during solidifying the liquid waste stream (calcination) and subsequent vitrification of the solid residu. Not all nuclides (e.g. Se-79, Ru-106, I-129) can be effectively fixed into a glass, since they hardly form stable compounds with the borosilicate matrix, or become volatile during the calcination process. Besides, still no safe final disposal facility for the highly radioactive glass is operational, only paper concepts exist

In spent fuel fission products and actinides constitute 3.5 and 1.4 mass% respectively. The solidified waste may contain up to about 30 % fission products plus actinides (IAEA-187 1979 [Q268]), so apparently a significant volume reduction might be achieved.

In this story the Zircalloy cladding hulls (0.6 - 2 Mg per Mg fuel) seem to be ignored, which also are highly radioactive with long-lived radionuclides.

Assumptions and fallacies

The waste volume reduction concept by vitrification, as communicated with the public, is based on starting points and assumptions which are questionable or even in conflict with basic laws of nature, such as:

- The view that short-lived radionuclides, which decay within a number of centuries (see Figure D2) would be not dangerous or less dangerous than long-lived.
- The view that waste containing less radioactive material per kg than spent fuel is nothing to worry about.
- The assumption that all long-lived radionuclides can be vitrified.
- The assumption that 100% complete separation of all chemical elements constituting spent fuel is feasible (see section D.2).
- The assumption that the borosilicate glass with the long-lived radionuclides will remain

stable for thousands of years and that no severe problems will arise with the borosilicate glass, caused by radiolytic reactions, heat generation, (re)crystallization and segregation of elements.

- Neglect of the substantial fraction of the fission products and actinides which are discharged into the environment by the reprocessing plant (see section D.2)
- Neglect of the large amounts of other radioactive wastes released during reprocessing (see Figure D1).
- Neglect of the immense amounts of radioactive waste resulting from decommissioning and dismantling of nuclear power plants and reprocessing plants.
- Neglect of an ever present consequence of the Second Law of thermodynamics: the unavoidable ageing and degradation of materials and structures, see Annex F.
- The assumption that waste containing short-lived radionuclides, however defined, could be stored safely in temporary above-ground facilities for more than four centuries and could be kept free of natural disasters, human failings, armed conflicts, criminal or terroristic actions.
- The assumption that future generations will have the economic means to maintain the storage facilities for the ‘short-lived’ radioactive waste adequately, without any revenue for them, as discussed in Chapter 9.
- The assumption that future generations will have the knowledge of the exact locations and properties of the stored ‘short-lived’ radioactive wastes from centuries before and will have the expertise and economic means to safely handle the wastes.

As pointed out above spent fuel contains many tens of different kinds of elements and only a limited number of these elements has the appropriate properties to be chemically immobilised.

Other waste streams

The nuclear industry does not discuss the large volumes of the remaining radioactive waste which cannot be vitrified, nor about the releases of large quantities of radionuclides into the biosphere. Nothing is said about the massive volumes of radioactive wastes resulting from the decommissioning and dismantling of the reprocessing plant at the end of its operational lifetime: immense amounts of debris and scrap (order of magnitude: a million of tonnes) are heavily contaminated with all kinds of radionuclides from the processed nuclear fuel.

The decay curves of Figure D2 show that after about 4 centuries the specific radioactivity of spent fuel is mainly set by the actinides. After about 1000 years the radioactivity of the fission products remains nearly constant at a level of slightly less than 1 GBq/kg for about 100000 years. This level is still more than a million times as high as the specific activity of the human body. Assume the ‘short-lived’ waste contains no actinides, accordingly to the advocated concept of the vitrification – which is not possible as explained in section D.2 –, which radionuclides are present in that waste?
In its communication with the public the nuclear industry seems to suggest that this level of specific activity is not harmful to humans anymore.

During the 1000 years before the specific activity of the waste reaches that ‘harmless’ level, the waste containers are not storaged in a deep geologic repository, according to the nuclear industry, but in above-ground facilities, which are less expensive. One can be sure that the waste containers will deteriorate by natural degrading processes, not counting human actions, to such an extent that much, if not all, radioactivity will end up in the environment.

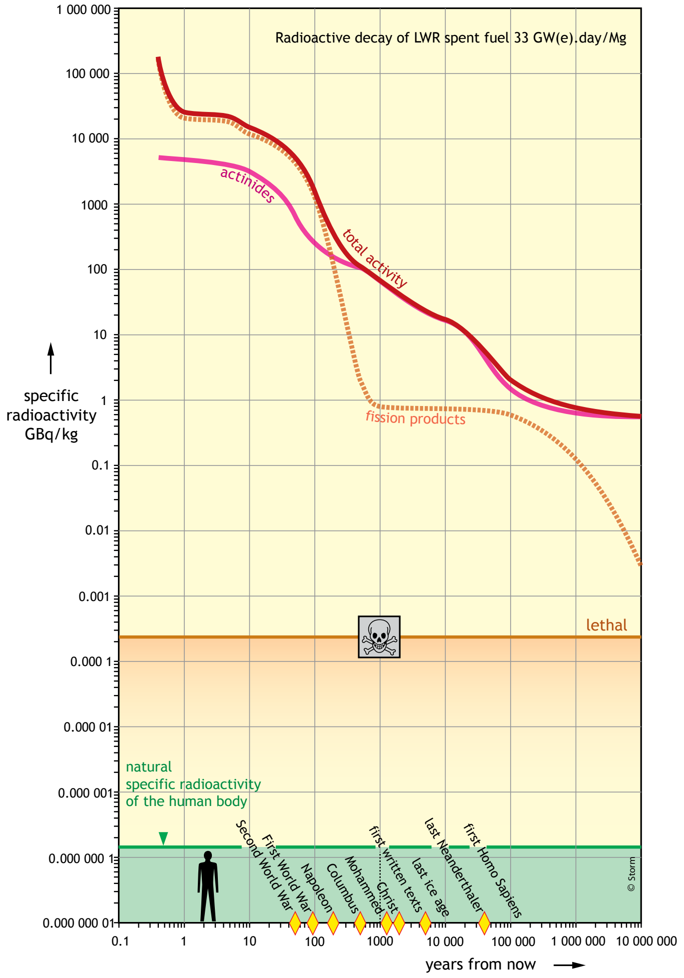

Figure D2

The specific radioactivity of spent fuel, measured in gigabecquerel per kilogram (GBq/kg). Nuclear fuel from the current types of nuclear reactors has considerably higher burnup than the fuel this diagram is based on and consequently has a higher specific radioactivity. The contributions of tritium, carbon-14 and activation products are not included in these curves. For more details see Annex A. The curves for all nuclear wastes together, including dismantling wastes, would be different from these curves.
Sources: Bell 1973 Q264 2], [3] Hollocher 1975 Q262, [4] JPL-77-69 1977 Q263, [17] Charpak & Garwin 2002 Q300.

Conclusion

Reprocessing offers no solution of radioactive waste problems. The amount of radioactivity in the nuclear waste streams is not influenced by the mechanical and chemical operations in a reprocessing plant. Discharging a significant part of the radioactive substances into the environment cannot be conceived as a ‘solution’ to the nuclear waste problem.
Actually the radioactivity from the spent fuel is dispersed over large volumes and masses of non-radioactive substances. Instead of volume reduction, the vitrification option results in a huge volume increase, coupled to a huge increase of the entropy ΔS of the radioactive components, exaggerating the waste problems beyond control.

The best way to handle spent fuel might be keeping the spent fuel elements intact, to pack them in very durable containers and to dispose of in a safe geological repository, as discussed in Chapter 2. Direct disposal poses the least risks and consumes the least materials and energy.

A number of countries, among other USA, Sweden and Canada, has chosen for this option. It is a fallacy to believe in ‘retrievable’ storage of spent fuel. In no way it is possible to extract net usable energy from it, when all industrial processes related to the vitrification of high-level waste, coupled to use of MOX fuel in LWRs, are accounted for.

One may wonder if the nuclear industry does propose reprocessing and vitrification as an excuse for not sequestering the dangerous radioactive waste permanently in a geologic repository. Apparently to justify the extremely high investments in the reprocessing industry, reprocessing is presently advocated by the nuclear industry as a means of reduction of the radioactive waste problem. This is a serious fallacy, as has been explained above.

D.6 Health hazards

Reprocessing poses serious health risks to the population of vast regions in several ways:
- routine discharges
- proliferation of plutonium
- ‘minor’ accidents, unnoticed or kept silent
- large accidents
- dismantling wastes

Routine discharges

A reprocessing plant ontaminates sea, air and land by its massive routine discharges with all kinds of radionuclides present in spent fuel, as has been pointed out in section D.3. The existence of these radioactive discharges are known, but not monitored by measurements by a genuinely independent institute.
A number of radionuclides are accumulating in the food chain, for example T, ^{14}C, ^{129}I, Pu, ^{90}Sr, ^{137}Cs. Especially seafood from the seas near the reprocessing plants (La Hague in Normandy, France, Sellafield in Cumbria in the United Kingdom) might be contaminated to such extent that consumption is not safe. The biological activities of these nuclides in the human body are poorly understood, let alone the activities of the many dozens of other radionuclides in the reprocessing waste. The health effects of the continuous radioactive discharges are not investigated.

Another impediment for sound health risks assessments is the fact that a number of dangerous radionuclides, e.g. tritium, carbon-14, iodine-129 and a number of alpha emitters, are hard to detect with commonly used radiation counters.
As a result of the difficult detectability, severe radioactive contamination with these radionuclides may escape notice during prolonged periods. Not every spill or release contains ‘marker’ nuclides which are easily detectable, such as ^{137}Cs.

Examples of ‘unnoticed’ releases are the routine releases of nuclear power plants under nominal conditions. For that reason it would be advisable to check on regular occasions food and drinking water in the vicinity of nuclear power plants on the presence of those troublesome radionuclides, even if no direct threat seems apparent.

‘Minor’ accidents

Unnoticed leaks are adding unknown amounts of radioactive materials to the discharges. No chemical plant is leakfree and reprocessing plants are very complex and very large. The functioning of these plants is shrouded in an aura of secrecy, so it may happen that 'minor' accidents, if noticed, are kept silent. Discharges by reprocessing plants into the environment are not monitored independently.

Large accidents

At a reprocessing plant immense amounts of radioactivity are stored, from many reactors from many years, probably in the order of 100000 nuclear bomb equivalents. A part of the high-leverl waste is stored in solid state, e.g. cladding hulls and insolubles, another part solved in liquids. Not known is to which extent the wastes are heat generating. By radiolysis of water hydrogen is formed, which has to be removed contantly. Buildup of considerable amounts of hydrogen is conceivable in reprocessing plants in case of accidents, major failures or human sloppiness, resulting in violent explosions. At high temperatures cladding hulls burn in air and react with water generating more hydrogen. Releases of massive amounts of radioactivity, equivalent with several reactor cores, cannot be ruled out.

Dismantling

Contamination is extensive in a reprocessing plant. Dismantling of the huge buildings will generate large volumes of heavily contaminated wastes. Costs will be high, because large parts of the construction are contaminated with dangerous long-lived radio-nuclides and alpha emitters. Experiences with dismantling the West Valley reprocessing plant and some minor DOE plants for processing nuclear materials, are not encouraging [83] UCS 2007 Q421 [49]n.

Dismantling of a reprocessing plant will be an exceedingly demanding task. Reprocessing plants are among the largest industrial complexes in the world. The hot areas, the compartments in which radioactive materials are processed, are strongly contaminated with radionuclides representing almost the entire Periodic Table of the Elements, including the transuranic actinides. The volume and mass of the radioactive debris and scrap resulting from the dismantling of a reprocessing plant will be a multiple of those from a nuclear power plant.

Unavoidably substantial amounts of radionuclides wil be released into the environment during cleanup and demolition of the buildings.
What will happen with the immense amounts of radioactive contaminated debris and scrap? Serious risks are posed by the poor controllability of the handling of the debris and scrap, illegal trade and criminality (see section 8.4) and the trend to relax the exposure standards when costs are mounting (see section 9.6).

D.7 Proliferation of plutonium and neptunium

By reprocessing of spent fuel large amounts of plutonium are produced. Use in MOX fuel poses serious risks of terroristic actions, as pointed out in section 6.2.

In 2005 a large leak (83 m^3) of a liquor containing dissolved spent fuel at the THORP (THermal Oxide Reprocessing Plant) reprocessing plant at Sellafield in the United Kingdom went undetected for more than eight months. The leaked solution contained some 19 Mg of uranium and 190 kg of plutonium and minor actinides. The fluids collected in a secondary containment. The fact that a shortfall in the amount of plutonium, enough for some 30 nuclear bombs, did not arouse concern for so many months, suggests that the theft of a significant amount of plutonium could also go undetected [52] Gronlund et al. 2007 [Q418 [43]n.

Neptunium-237 is fissile as are the isotopes of americium (^{241}Am, ^{242m}Am and ^{243}Am), so these radionuclides are usable for nuclear weapons. Neptunium can be relatively easily separated from uranium and plutonium because of it is a distinct element with its own specific chemical properties. In a reprocessing plant pure neptunium can be produced from spent fuel and due to the long halflife of ^{237}Np (2.14 million years) its the only isotope recovered. The other isotope ^{239}Np has a halflife of 2.36 days and disappears almost entirely from spent fuel by decay to ^{239}Pu within weeks after shutdown of the nuclear reactor. In this way a pure fissile material can be obtained by means of chemical processes. Plutonium is always a mixture of isotopes, a part of which are not fissile.

The production of americium and the handling of the material are much more demanding than of neptunium.

Neptunium and americium are outside international control and are not safegarded by IAEA (ISIS 1999 Q552).

D.8 Summary

Reprocessing of spent fuel is a sequence of separation processes, involving chemical equilibria which are governed by fundamental laws of nature, especially the Second Law of thermodynamics. From the Second Law follows that separation of a mixture of a number of chemical species into an equal number of 100% pure fractions is impossible. Consequences are:
- all fractions (uranium, plutonium, fission products and minor actinides) will be contaminated with traces of the other fractions,
- unavoidably a part of all fractions will be lost in the waste streams,
- unavoidably a part of all fractions will be discharged into the environment,
- it is not possible to produce 100% pure materials, with perfectly predictable properties.

Separation of a mixture into fractions becomes more difficult and less complete as:
- more different species are present in the mixture
- the concentration of the desired species is/are lower
- the species in the mixture are chemically more alike
- the radiation levels in the mixture (solution) are higher.

More difficult means the necessity to invest more energy, chemicals and equipment to attain an acceptable level of separation, and consequently higher costs.
Purity of materials is often an economic notion.

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ANNEX **E**

Partitioning & transmutation

E.1 Concept and its promises

The activity of the fission products in spent fuel falls with a factor of one million during the first four centuries after discharging from the reactor, as Figure D2 (see Annex D) shows, and the activity of the actinides with a factor of ten thousand. After that period, the radioactivity decreases very slowly.

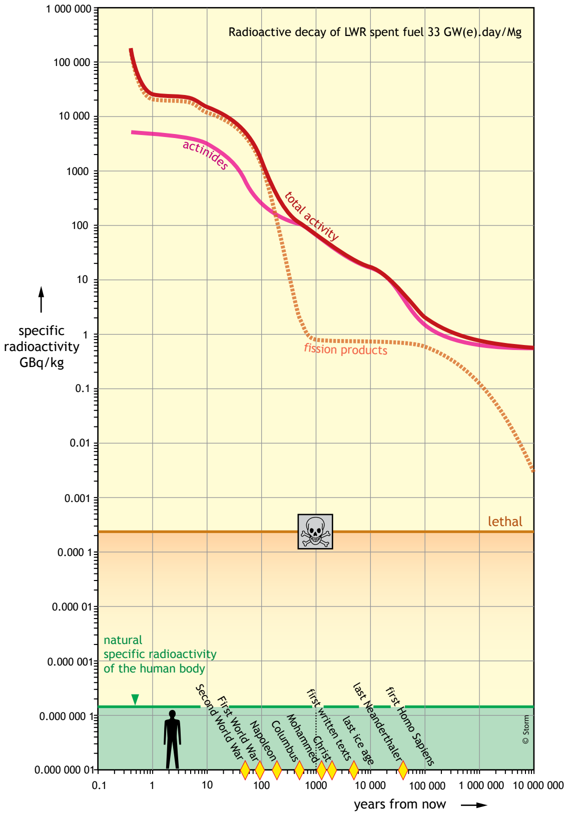

Figure E1

The specific radioactivity of spent fuel, measured in gigabecquerel per kilogram (GBq/kg). Nuclear fuel from the current types of nuclear reactors has considerably higher burnup than the fuel this diagram is based on and consequently has a higher specific radioactivity. The contributions of tritium, carbon-14 and activation products are not included in these curves. This diagram is identical to Figure D2 in Annex D and Figure A6 in Annex A.

Sources: Bell 1973 Q264 2], [3] Hollocher 1975 Q262, [4] JPL-77-69 1977 Q263, [17] Charpak & Garwin 2002 Q300.

Theoretically it is possible to convert long-lived radionuclides into short-lived ones by neutron irradiation. This conversion is called transmutation, because the long-lived radionuclides belong to other chemical elements than those resulting from the neutron irradiation. The term transmutation originates from the Middle Ages when alchemists tried to transmutate lead into gold. Transmutation by chemical means is not possible.

Partitioning & transmutation is advocated by the nuclear industry as a means of ‘destroying’

long-lived radionuclides. Were it possible to convert the long-lived nuclides into short-lived or stable ones, the concentrations of the long-lived dangerous radionuclides in the remaining wastes could be reduced to below an official standard, so it would be safe for release the waste into the public domain after a storage time of 'only a few' centuries to a thousand years, instead of hundreds of thousands of years.
In such way the amount of high level waste to be stored permanently in a geologic repository could be reduced to a small fraction of the spent fuel. See also for example CEA 2002 Q448, DOE-NE 2009 Q450, NWMO 2008 Q446, ORNL 2011 Q449, NRC 1996 Q16 and SKB 2010 Q447.

Shifting focus

During the early 1990s P&T came into the spotlight of the nuclear world. From a political viewpoint the P&T concept was very attractive for it was advertised as the road to nuclear energy producing much less radioactive waste than the current technology, waste that would remain dangerous for only a couple of centuries. In addition the existing radioactive wastes could be reduced to little problematical proportions. Some publicaties suggest even the possibility of nuclear energy with (almost) no radioactive waste (LPSC 2001 Q451, ORNL 2013 Q539).

At first the research focused on transmutation by means of existing reactors (LWR) and by yet-to-be-developed Accelerator-Driven System (ADS), see section E###. During the past decade the focus of the research shifted to the development of fast reactors of the so-called Generation IV class and the treatment of the minor actinides (MA). Transmutation of long-lived fission products, such as ^{99}Tc and ^{129}I, seems to be pushed tacitly into the background of the current investigations. On closer consideration this shift turns out to be caused by the choice for fast reactors to be used in a proposed P&T system: this type of reactor is not suitable for transmutation of long-lived fission products.

The envisioned advanced fast reactors of Generation IV – called 'fast' because they operate with fast neutrons – would have the advantages of the intended breeder reactors from the 1970s and 1980s plus the ability to fission the minor actinides. On the one hand these reactors would be able to generate a 100 times more energy from a given mass of natural uranium than the currently operational power reactors, on the other hand they would greatly reduce the radioactive waste problem.
Evidently this sounds as a polically attractive concept. In addition P&T research may contribute to fundamental physical and chemical knowledge and offers an argument to postpone the politically very uninviting investments in the isolation of radioactive waste in a geologic repository. This postponement fits very well the current paradigm *après nous le déluge* and living on credit

E.2 Transmutation

Two nuclear processes are important in a P&T system:
- Transmutation in the classic sense
- fission of actinides

Transmutation

By neutron capture and subsequent radioactive decay of the nucleus an atom can transmutate into an atom of another chemical element.Depending on the characteristics of the nucleus that captures a neutron, different outcomes are possible:

- A stable nucleus transforms into another stable nucleus of the same element, so no transmutation occurs.
- A stable nucleus transforms into an unstable nucleus which spontaneously transmutates by radioactive decay into the nucleus of another element, with short or long half-lifes, depending on the involved nucleus. This process, usually coined activation, results in an increase of the radioactivity of the mixture
- An unstable nucleus transforms into another unstable nucleus of the same element; the new radioisotope generally has a shorter half-life than its parent.

The second process causes an increase of the radioactivity of the irradiated mass and is undesired.
The third process is aimed for in the P&T concept: the transmutation of long-lived radionuclides into short-lived ones, which will decay into stable nuclides within a 'short' time (however defined). For most long-lived radionuclides such a nuclear transmutation process is possible, in principle, but not for all.
Because neutron irradiation occurs randomly and cannot be aimed at specific nuclides in a mixture, all processes occur simultaneously. The only way to prevent unwanted nuclear reactions is to remove the involved nuclides from the mixture by means of chemical separation processes.

As a consequence of the transmutation of long-lived radionuclides into short-lived ones the specific radioactivity of the irradiated waste rises sharply.

Fission of actinides

Fission of actinides, notably: neptunium (Np), plutonium (Pu), americium (Am), curium (Cm), berkelium (Bk) and californium (Cf). These nuclides, often called transuranics or trans-uranium elements, with the acronym TRU, are generally considered to be the most dangerous components of spent fuel. By fission of a heavy nucleus two or three lighter nuclei of other chemical elements come into being.
A part of these fission products are radioactive with long half-lifes.

E.3 The system

A partitioning & transmutation system consists of a transmutation cycle and a waste management system. The cycle consists of three components (see Figure E1):
- partitioning
- fuel and target fabrication
- transmuter.

Each of the three components must operate flawlessly and finely tuned to the other two components in order to attain an appreciable transmutation rate. If one component fails, the whole system fails.
The cycle starts with partitioning of spent fuel into a number of fractions. The fractions to be transmuted are concentrated into special fuel elements, targets, which are placed into the transmuter reactor. In the core of the reactor conventional fuel elements are placed to maintain the fission process, which is the source of neutron radiation.
After discharging from the transmuter, the irradiated fuel and targets must be reprocessed. The remaining long-lived nuclides must be separated from the stable and short-lived nuclides, and processed into new transmutation targets for the next irradiating cycle.

Inevitably, in a transmuter reactor nonradioactive atoms capture neutrons as well, becoming radioactive. Also, a part of the uranium in the reactor is converted into new trans-uranium actinides by neutron capture. By fission of actinides (TRUs) new long-lived fission products are generated.

During the irradiating time in the reactor, only a small part of the unwanted nuclides is transmuted, while new ones come into being by above mentioned processes. Therefore, the net transmutation rate is lower: for many nuclides not more than a few percents per cycle. To attain a significant reduction in the inventory of unwanted long-lived radionuclides, many cycles are needed.

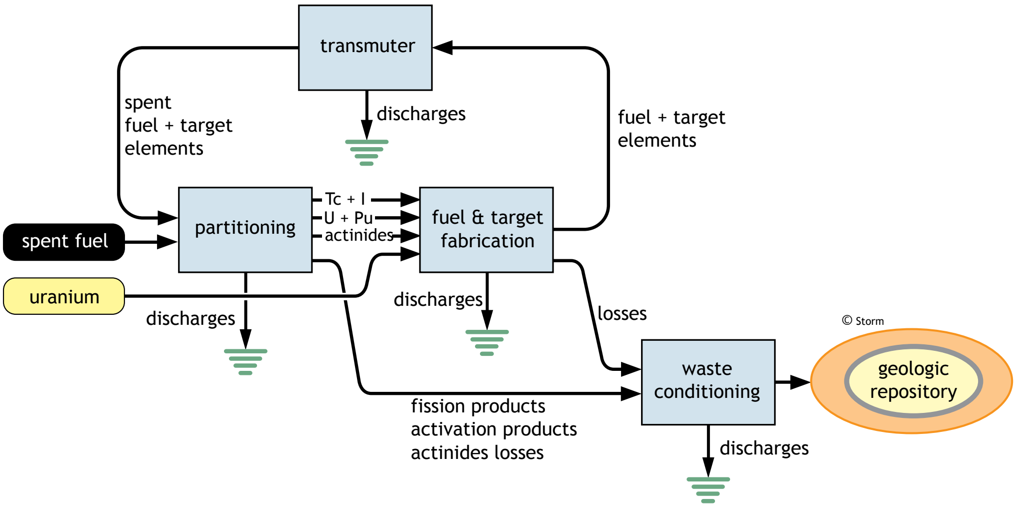

Figure E1
Flows of radioactive materials in the proposed concept of a partitioning & transmutation system. The system consists of a transmutation cylce plus a waste conditioning facility and a geologic repository, Three components form the cycle: a transmuter (reactor), a partitioning (reprocessing) plant and a facility to produce fuel elements and targets containing the nuclides to be transmuted. The radioactive wastes from the cycle are to be conditioned and stored in geologic repository. In this flowchart the inputs of chemicals, materials and energy have been omitted, as well as the waste streams arising from decommissioning and dismantling the facilities.

E.4 Partitioning

Outline

Spent fuel contains a mixture of many dozens of different kinds of atoms (chemical elements), often with different isotopes. This exceedingly complicated mixture has to be sorted into several fractions in a process called partitioning, a crucial step in the P&T cycle.
The separation processes are based on the differences between the chemical properties of the constituents of the mixture. It is not possible to sort stable and radioactive isotopes of the same element, nor is sorting according to half-lifes. Atoms of some elements are chemically very similar and consequently are difficult to separate.
For example the lanthanides have similar chemical properties as the actinides, but they have to be separated from them. Most lanthanides have stable nuclei and transform into other lanthanides by neutron capture. More important is that the lanthanides exhibit strong neutron absorbing properties to such extent that the transmutation of the actinides is hampered by their presence in a transmuter.

Generally the chemical treatment of spent fuel is called reprocessing. A greater number of fractions is the only difference between partitioning and reprocessing. Partitioning would be a more complicated version of the reprocessing of spent fuel from closed-cycle fast reactors, which in turn would be an advanced and much more complicated version of reprocessing of LWR spent fuel. Reprocessing is also a crucial step in the realisation of the closed-cycle reactor.

Uranium and plutonium are recycled to produce new fuel elements for the transmuter reactor needed to generate the neutrons required for the transmutation processes. The remaining and newly formed actinides have to be separated from the spent fuel and target elements to be reworked into new target elements. Short-lived and stable fission products are to be separated and packed into waste containers. Nuclides, albeit long-lived radioactive or stable, which poison the fission and transmutation processes in the transmuter are to be removed from the mixture and from the cycle.

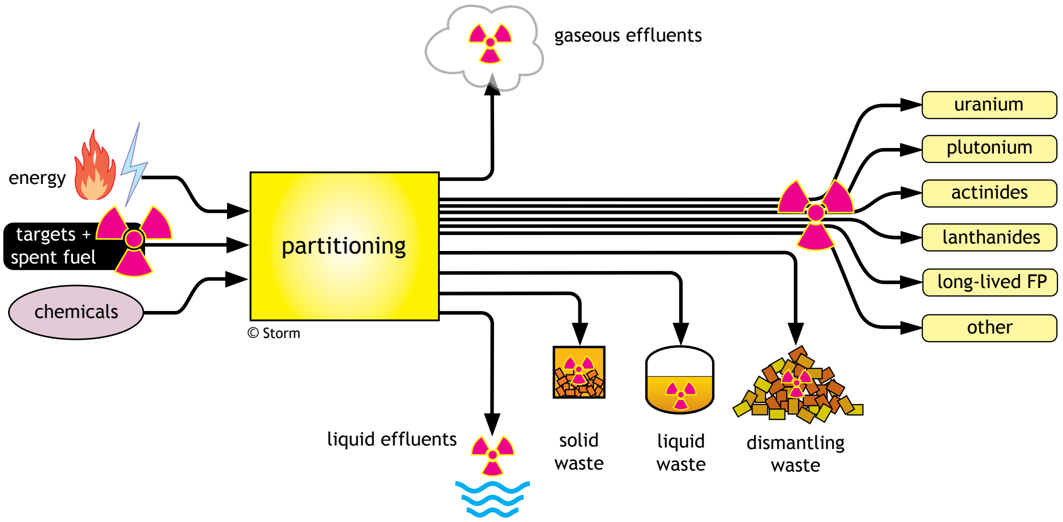

Figure E2
Outline of the partitioning of fuel and target elements of a transmutation cycle. For details see text.

Difficulties

By recycling, the composition of plutonium shifts like that of uranium. The amounts of trans-plutonium elements increase with each reprocessing cycle. Due to this, the alpha-, gamma- and neutron radiation rise (with a factor 3), as well as the specific heat generation of the plutonium by radioactive decay with a factor 7 (ORNL-TM-2879 1970 [Q254], Fischer 1986 [Q240] and Roepenack et al. 1987 [Q241]). Evidently these facts cause mounting difficulties in the handling of the recycled uranium and plutonium.
Some isotopes have a very low critical mass for a fission chain reaction. Serious criticality problems complicate the reprocessing of fuel with high trans-plutonium content. For example, the critical mass of Am-242m may be as low as 7 grams, according to Ronen et al. 2000 [Q243].

Besides the rising radioactivity, the proportion of fissile isotopes declines each time the plutonium is recycled. Both effects cause a rapid deterioration of the practical use of recycled plutonium.

Reprocessing of AFR fuel, which would have a high burnup (typically 70-100 GW(th).day/Mg) is more troublesome than of LWR fuel, with a burnup of 30-50 GW(th).day/Mg. Burnup is a measure of the stay time in the reactor and of the neutron flux the fuel gets in the reactor. The higher the burnup, the more atoms per Mg fuel are fissioned and the higher is the neutron flux. This results in higher concentrations of plutonium, in more trans-plutonium actinides and in larger specific amounts of fission products than in LWR fuel. Spent fuel from an AFR or a transmuter reactor is much stronger radioactive than spent LWR fuel.

High burnup and the high Pu content have some difficult consequences for the dissolving and separation processes in the reprocessing plant (UNIPEDE/CEC 1981 [Q58]):

- Certain reactions are no longer of a secondary significance, as in LWR fuels, e.g. radiolysis, precipitation, insolubility and corrosion. A high metallic fission product concentration increases the proportion of insoluble material, which may trap plutonium and has a high heat and radiation output.
- Higher plutonium concentration entails a lower dissolution rate and the formation of insoluble compounds in significant quantities.
- Higher specific radioactivity causes more radiolyse of the extraction liquid, an organic solvent (tributylposphate TBP). Solvent degradation products may clog pipelines and extractors, thus blocking the entire process.
- Plutonium may accumulate by formation of complex compounds with solvent degradation products, enhancing the risk of criticality accidents.
- Some isotopes of the minor actinides have a very low critical mass for a fission chain reaction. Serious criticality problems complicate the reprocessing of fuel with high trans-plutonium content. For example, the critical mass of Am-242m may be as low as 7 grams, according to Ronen et al. 2000 [Q243].
- More insoluble compounds and noble metal alloys (Ru, Rh, Tc, Mo, Pd), possibly containing plutonium, and undissolved MOX particles remain in the dissolver. The undissoved particles cause high plutonium losses, solvent degradation by high heat and radiation emission and plugging of lines and equipment.
- Higher specific heat generation demands more elaborate temperature control.
- The high plutonium-uranium ratio and the presence of large specific amounts of fission products make the extraction process more complex and less efficient. Short contact times between the organic and aqueous phase might be required, even if this feature reduces the effectiviness of plutonium separation. More U and Pu is lost in the waste streams and the U and Pu product stream is more contaminated with actinides and fission products.
- Higher concentrations of plutonium and other actinides enhance criticality problems in the separation system.This necessesitates a design combining safe geometry and proper monitoring. To overcome geometry constraints a modular design may be needed. In that case a reprocessing plant in an AFR or P&T cycle would benefit less from advantage of scale than a LWR fuel reprocessing plant.

Some other difficulties for spent AFR fuel and even more for spent fuel and target elements from a transmuter reactor, caused by the high radioactivity and heat output of the irradiated materials, are:

- The fuel assemblies of a AFR, made of stainless steel, have to be disassembled before the fuel pins can be chopped into smalle pieces.Due to dimensional changes as result of the fast neutron irradiation, this part of the process will be difficult.
- Keeping out-of-pile time as short as possible, needed for a high plutonium gain in the cycle, enhances problems with transport and handling of the irradiated fuel elements.
- As result of the short cooling time after removing from the reactor core – preferable less than 1 year versus LWR fuel cooling times of 3 years or more – the shearing of the fuel elements releases gaseous highly active short living fission products, such as I-131.

The effluent release after short cooling times causes two main problems. First, the higher decontamination factors required on gaseous effluent for reduction to acceptable operational release levels. Second, the higher potential risk of accidental release and the necessity of increased engineered safety measures, to reduce the overall risk to acceptable levels.

- The inevitable discharges into the environment of actinides will be higher than in a LWR cycle, because of the higher actinide content in AFR and transmuter fuel and target elements.

E.5 Fuel element and target fabrication

A transmuter reactor has to be fuelled with uranium-plutonium fuel elements to maintain a fission process which would generate the neutrons needed for the transmutation. The radionuclides to be irradiated and transmuted have to be incorporated into special target elements, which are also placed into the transmuter reactor. These target elements have to comply with very stringent nuclear physical conditions, in order to make the transmutation process as efficient and as safe as possible.

Due to the very high radioactivity, including neutron radiation, the fabrication plants has to operate remotely controlled. The high radiation levels might pose serious problems even for robotic equipment, especially the electronics.

E.6 Transmuters

Three principal transmuter concepts are based on respectively a thermal-neutron reactor, a fast-neutron reactor and an accelerator-driven subcritical nuclear reactor. As thermal reactor the light water reacor (LWR), PWR or its Russian counterpart, is proposed. The fast reactor should be the advanced liquid metal reactor (ALMR), sodium- or lead-cooled. Several variants of the accelerator-driven transmuter are proposed, e.g. ATW (Accelerator Transmutation of Waste) and Phoenix in the USA and the ADS-800 in Russia (NRC 1996 [Q16], Bergelson et al. 2002 [Q50]). A fourth transmuter concept is the PBR (particle bed reactor or pebble bed reactor), a helium-cooled fast reactor.

In an elaborate study (NRC 1996 [Q16]) three transmuter concepts have been compared. The PBR and Phoenix proposals did not have sufficient detail for an adequate comparison with the principal three, to be discussed below.

LWR

Current LWR designs could be modified for transmutation of most actinides and the more troublesome fission products, e.g. technetium and iodine. TRUs could be recycled in MOX fuel in about a third or more of the reactor cores. In burning TRUs, however, thermal reactors like the LWR tend to build up ^{240}Pu, ^{241}Pu, ^{242}Pu and higher actinides: Am, Cm, Bk and Cf. These affect reactivity in the reactor core and are a severe complication for reprocessing and recycle fuel fabrication and quality assurance.

ATW

The designers of the ATW (Accelerator Transmutation of Waste), also called Accelerator-Driven System (ADS), propose either aqueous slurry (in D_2O) or molten salt systems. The ATW system includes a large proton accelerator (a machine nearly 2 kilometers long), a beam target of tunsten or molten lead on which the protons are focussed to generate high-energy neutrons, an 8000 MW(th) subcritical reactor, an on-line reprocessing system, and the operation of a 1600 MW(e) power plant to power the proton accelerator. To achieve reasonable on-line availability of such a system would be a major challenge.

The ATW poses major engineering and materials challenges due to the extraordinary operating conditions in the reactor and beam target. Heat removal is a major issue for development as well as for safety and licensing. Also, the possibility of reactor transients, unevaluated in any detail at present, may require means of reactivity control beyond that afforded by merely turning off the accelerator.
The ATW uses fluid fuels, which involve unproved technologies for fuel fabrication and on-site reprocessing and which raise a number of operational and safety issues for which detailed analyses have not yet been done.
The ATW would produce more higher actinides (TRUs) than an LWR.

ALMR
The ALMR, Advanced Liquid Metal Reactor, under development for decades as a fast breeder reactor could be modified to operate as a TRU (transuranics) transmuter. For the ALMR, the limiting breeding ratio is 0.65; below this, there may be a safety problems because of a positive void reactivity coefficient. A low value is required for actinide burning. At a breeding ratio of 0.65 the ALMR would be comparable to an LWR in net TRU burning rate. Because of tits fast-neutron spectrum the ALMR would have impractically low transmutation rates for long-living fission products.

AFR and IFR

During the past decade the focus of the research shifted to the development of fast reactors of the so-called Generation IV class and the treatment of the minor actinides (MA), as pointed out in section E.1. Several designs are being investigated: cooled by liquid sodium, liquid lead, molten salts or other, sometimes called AFR, for Advanced Fast Reactor, or IFR, for Integrated Fast Reactor. These reactors can burn actinides, but are not suitable for transmutation of long-lived fission products, as pointed out above.

The envisioned advanced fast reactors of Generation IV – called ‘fast’ because they operate with fast neutrons – would have the advantages of the intended breeder reactors from the 1970s and 1980s plus the ability to fission the minor actinides. On the one hand these reactors would be able to generate a 100 times more energy from a given mass of natural uranium than the currently operational power reactors, on the other hand they would greatly reduce the radioactive waste problem, according to the nuclear industry.

The term ‘breeder’ originates from the feature of this type reactor that it could breed more fissile nuclei than it consumed. In such a fast reactor – fast because it operates with fast neutrons, not because it would breed fast – more fissile plutonium nuclei would be formed from non-fissionable uranium-238 than it would fission. The breeder concept promised to be able to fission about 60% of the nuclei in natural uranium. In conventional reactors, mostly LWRs, not more than about 0.6% of the nuclei can be fissioned. The breeder concept promised to generate 100 times more energy from a given mass of uranium than conventional reactors.

Of the advanced fast reactors of Generation IV the liquid sodium-cooled fast reactor has reached the farthest stage of development and is a direct offspring of the LMFBR (Liquid Metal-cooled Fast Breeder Reactor). The LMFBR, in effect the breeder cycle, has been under development for more than five decades in a number countries, e.g. USA, UK, France, Germany, former USSR now Russia, Japan and India. In the USA and Europe alone the investments amounted to some €100bn. These efforts so far have failed to demonstrate that the breeder cycle (^{238}U-^{239}Pu) is technically feasible. This failure is not a matter of advanced technology and steady development, the problems preventing the realisation of a closed-cycle reactor functioning according to the promises are of fundamental sort. The concepts of closed-cycle reactors, the ^{238}U-^{239}Pu breeder as well as the ^{232}Th-^{233}U breeder, are implicitely based on the assumption that perfect materials, perfect equipment and perfect separation processes could become available. Exactly these conditions are impossible, as a consequence of the Second Law of thermodynamics, as will be explained in Annex F.

The nuclear industry attributes the stagnation of the research on the LMFBR to bad economics in a time of cheap uranium and also to opposition from environmental groups. Politically it became less attractive to award large sums to the development of the LMFBR. With the introduction of the P&T concept the old LMFBR concept revived under new names, such as: advanced closed-cycle reactor, AFR, IFR, ALMR, Generation IV. The Generation IV class comprises also other old (40-50 years) concepts, which never came to maturity, such as molten-salt reactors (e.g. MSFR, Molten Salt Fast Reactor), helium-cooled fast reactors, lead-cooled reactors and thorium-fuelled reactors.

E.7 U and Pu in closed-cycle reactors

Reprocessed uranium

Recycled uranium, from reprocessed spent fuel (see also Annex D) also called reprocessed uranium U(rep) has a different composition than natural uranium and contains a number of nuclides which degrade its properties as nuclear fuel (Forsey & Dickson 1987 [Q239]) such as:

- uranium-232,decaying to thallium-208 with high alpha and gamma activity
- uranium-234, a strong alpha emitter
- uranium-236, a strong neutron absorber; fuel with this isotope needs a higher enrichment assay or more fissionable plutonium to compensate for it,
- traces of fission products, like ruthenium-106 and technetium-99, which increase the gamma activity of the uranium
- traces of trans-uranium elements, e.g. neptunium and plutonium.

The gamma activity of recycled uranium increases with time, because of a growing content of gamma-emitting decay products.

Recycled uranium needs more enrichment, or addition of more fissile plutonium, than depleted or natural uranium, because of the neutron absorbing properties of ^{232}U, ^{234}U and ^{236}U. These even-numbered isotopes of uranium are not fissile. Enrichment of recycled uranium by gasdiffusion or ultracentrifuge is questionable, because the uranium is enriched more effectively in the two lightest isotopes ^{232}U and ^{234}U than in ^{235}U and is enriched also in ^{236}U, to lesser extent, making the enriched product strongly radioactive. The reenriched uranium would contain the largest part of the unwanted uranium isotopes and some other unwanted nuclides mentioned above as well. Besides, the enrichment plant would be contaminated with highly radioactive compounds, hampering its operation.

For above reasons reprocessed uranium has not been used in power reactors. The global inventory may be some 25000-30000 Mg U(rep) and stored in a number of facilities. Evidently this material poses health risks when released into the public domain, by accidents, terroristic actions or otherwise.

Plutonium

Reactor-grade plutonium originates from spent fuel from civil power reactors and contains typically less than 70% fissile plutonium isotopes (^{239}Pu + ^{241}Pu). In these reactors the fuel elements stay far longer an get a higher burnup (33000-46000 MW(th).days/Mg). Due to the long stay time in the reactor, more of the heavier plutonium isotopes are formed: ^{240}Pu, ^{241}Pu and ^{242}Pu, but also ^{238}Pu. The even isotopes are not fissile in LWR's and in bombs. Moreover, trans-plutonium elements, e.g. ^{241}Am, ^{243}Am, ^{244}Cm, are formed from plutonium isotopes by neutron capture.

Pu-238 is a strong alpha emitter. By beta decay, plutonium-241 is transformed into americium-241; ^{241}Am is a strong gamma emitter, greatly increasing the gamma activity of the plutonium. Within a few years storage time, the concentration of ^{241}Am builds up to a level the plutonium cannot be handled safely anymore. With a content of ^{241}Am higher than 1% it has to be purified again (Hulst & Mostert 1979 [Q242]), a very costly process. For recycled plutonium from LWR with MOX fuel, the repurifying limit due to ^{241}Am may be reached about one year after reprocessing.

Most plutonium and trans-plutonium isotopes emit neutrons, as some of their nuclei spontaneously fission (the other nuclei decay by alpha or beta emission). The presence of all these nuclides makes reactor-grade plutonium a hazardous substance, with troublesome properties as reactor fuel. The problems with increasing gamma, alpha and neutron radiation aggravate with each recycling of the plutonium.

By recycling, the composition of plutonium shifts like that of uranium. The amounts of trans-plutonium elements increase with each reprocessing cycle. Due to this, the alpha-, gamma- and neutron radiation rise (with a factor 3), as well as the specific heat generation of the plutonium by radioactive decay with a factor 7 (ORNL-TM-2879 1970 [Q254], Fischer 1986 [Q240] and Roepenack et al. 1987 [Q241]). Some isotopes have a very low critical mass for a fission chain reaction, e.g. Am-242m (maybe as low as 7 grams, according to Ronen et al. 2000 [Q243]), complicating the reprocessing of fuel with high trans-plutonium content.

E.8 Feasibility of P&T systems

Besides the limitations set by the Second Law of thermodynamics, other issues should be considered: when assessing the feasibility of a partitioning & transmutation system which meets its promises:
- timeframe
- inherent flaws
- technical imperfections
- integration
- total amount of radioactive waste
- impact on the net energy production of a nuclear energy system as a whole.
- costs
- human resources

Timeframe

According to the Swedish study SKB 2010 Q447 the development of a functioning p7t system would take several decades. In view of the experiences in the past with the development of the U-Pu breeder cycle and the fact that a P&T cycle is more demanding than the U-Pu breeder cycle, this estimate might be optimistic.

Even in case of a flawlessly operating system according to the design specifications, extremely long transmutation operating times for TRUs are needed. Assuming a constant level of nuclear power in the future, a transmutation time of a thousand years would be required to reach a hundredfold reduction in TRUs inventory using the accelerator-driven transmuter and many thousands of years using an LWR or ALMR transmuter. This time could be reduced to a few centuries if nuclear power were to be terminated as rapidly as possible (NRC 1996 [Q16]).

According to SKB 2010 Q447 the perfect functioning of a number of P&T systems during at least one century and probably longer will be necessary to fission the existing amounts of actinides.

Two hypothetical examples may make clear the needed operating time to reduce a given amount of long-lived radionuclides to 1% of the original value. Both computational examples are based on data from NRC 1996 [Q16] and assume flawless operation of the P&T system, without process losses in the P&T cycle.

1 In an ADS operating with thermal neutrons 2.5-3% of the long-lived fission products could be transmuted per cycle into short-lived radionuclides. Assuming a net transmutation rate of 3% per cycle, 152 cycles would be required to transform 100 kg long-lived radionuclides into 99 kg short-lived or stable nuclides and 1% remaining long-lived radionuclides. Assuming a cycle time of one year, an uninterupted period of 152 years would be required.
   Transmution rate of actinides with thermal neutrons would be not more than 2.2% per cycle.
2 An ADS operating with fast neutrons the transmutation rate for actinides could be 22% per cycle. In this case 18 cycles would be sufficient for a reduction to 1% of the original amount, corresponding with 18 years assumed the cycle time is 1 year.
   Long-lived fission products are not or barely transmuted by fast neutrons.

A cycle time of 1 year is unlikely short in view of the extreme complexity of the of the processes in the cycle and the high radioactivity of the materials. Experiences with current reprocessing plants, with their much less demanding tasks, point to cycle times of 10-20 years. Such long times would result in 1500-3000 years in example 1 and 180-360 years in example 2.

Inherent flaws

With regard to the transmutation of long-lived fission products and activation products the proposed P&T systems, even if functioning at design specifications, show inherent nuclear flaws, among other:

- Transmutation of ^{99}Tc and ^{129}I, two long-lived fission products of major health risk concern, is marginal at best, even under design conditions.
- Transmutation of ^{14}C, another troublesome long-lived radionuclide in nuclear waste, is not possible, because of its exceedingly small neutron cross section.
- Transmutation of ^{90}Sr and ^{137}Cs, of major concern during the first thousand years of waste storage, is not practical, as little as transmutation of the long-lived nuclide ^{135}Cs.
- Transmutation of other fission products (e.g. ^{79}Se, ^{93}Zr, ^{107}Pd, ^{126}Sn, ^{151}Sm) is not included in the studies, nor are long-lived activation products (e.g. ^{59}Ni, ^{93}Mo, ^{94}Nb).
- Transmutation of TRUs in wastes never can be complete: theoretically at best a reduction to about 1% of the original amount may be achieved. In practice the reduction will be much less effective, for nu technical system operates perfectly, let alone during its whole operational lifetime.

These facts may be the reason why the nuclear industry shifted its focus from transmution of all long-lived radionuclides to the fission of actinides only. In many publications to the public the nuclear industry makes this limitation not very clear.

Technical challenges

Because the nuclides to be transmuted must be recycled many times through the transmutation cycle, the separation processes have to meet very strict requirements. The (unavoidable) process losses should be lower with a factor 1000 or more than achieved with current technology. It has not been proved that requirements could be met.
Complete separation of a mixture into 100% pure fractions is not possible. This observation follows from the Second Law of thermodynamics, as is explained in section D.2.

Considerable R&D would be required to assure that the low breeding ratio of a burner would not create safety problems and to assure that changes in fuel composition caused by burn-up would not adversely affect performance and safety. The ALMR as part of any Integral Fast Reactor (IRF) design includes pyroprocessing of its spent fuel. This technology is still in an early stage of development and its feasibility has not been proved.

The accelerator-driven transmuter concept, consisting of a suncritical reactor coupled to a particle accelertor, is based on unproven technology. Extensive research and development would be required even to ascertain wether an ATW system (Accelerator Transmutation of Waste) is technically feasible (NRC 1996 [Q16]).

Fabrication of fuel and target elements will pose a major challenge, because of the high concentrations of TRUs and other radioactive nuclides. The facility has to be operated remotely. Even with robots the fabrication process would be a major challenge.

The three main components of a P&T system – partitioning, fuel + target fabrication and transmuter – would have to be able to operate during many decades continuously and flawlessly.

Integration

Effective transmutation not only depends on a well-operating transmuter, but also on flawlessly operating separations and fuel plus target fabrication facilities. In addition the three components have to be finely tuned to each other and have to be integrated into one system. Materials in storage or in transit are not irradiated, slowing down the transmutation rate. Stay times of the materials outside the transmuter should be kept to a minimum, which requires a far-reaching integration of the partial systems, a daunting task (NRC 1996 Q16).

The failure of the integration of reprocessing, fuel fabrication and fast reactor into a functioning breeder cycle is an important cause of the proved unfeasibility of the breeder cycle, apart from the unsurmountable problems with each of the partial systems. Essentially these problems stem from fundamental limitations following from the Second Law of thermodynamics (see Annex F).

Energy balance

Transmutation is proposed as a waste reduction system as part of an energy-generating system, not as a goal in itself. In judging transmutation on its merits, energy consumption of the system as a whole should be an important parameter. Before embarking on the development of a P&T system, first the energy balance of the whole complex of power reactors and waste handling

should be drawed up.
All activities, like construction and operation of the facilities of the partition & transmutation cycle and all waste streams, including dismantling wastes of all facilities of the cycle, should be accounted for. A geologic repository would still be needed, for disposal of long-lived fission products and activation products, including dismantling waste.
As pointed out in Annex D, reprocessing is a very energy-intensive process and the energy consumption of the dismantling of reprocessing plants will be exceedingy high.

Costs

According to SKB 2010 Q447 it is not possible in the present stage of development to estimate the cost of a P&T system reliably in any way. Experiences in the past with development of the U-Pu breeder cycle are not encouraging. History shows that the preliminary cost estimates of large complicated technical systems are always too low, often by a factor 2-5 but not seldom by a larger factor (RAND 1979, RAND 1981). This phenomenon is not limited to nuclear projects.

Cost overruns are the rule in the nuclear technology. Even the construction cost of conventional nuclear power stations (e.g. Olkiluoto, Flamanville) seem to be hardly controllable. The factors causing the excessive cost escalations of large new projects as identified by the RAND studies perfectly apply to the development of a P&T system. As the first indications of extremely high costs of a P&T system may serve the first rough estimates of the dismantling cost of conventional reprocessing plants: some €10 million per tonne reprocessed spent fuel.

Human resources

A P&T system has to operate for at least several centuries to treat the waste from a given reactor after closedown of that reactor. Great numbers of highly skilled people will be needed to perform these demanding processes. Even these days the nuclear industry has difficulties to recruit and to educate enough highly skilled workers to sustain and operate its facilities and nuclear power plants (SKB 2010 Q447).

Massive amounts of radioactive materials are to be processed after closedown of a nuclear power plant, a part of which is highly radioactive. Even if the last nuclear power plant would be shut down today, the economy has to sustain a nuclear workforce for centuries to come. This workforce will not contribute to any improvement of the energy supply. Its sole task is to prevent the nuclear legacy from going disastrous.
One may wonder if enough young people would opt for the required rigorous education and training and if a free market-oriented economy would easily support such a workforce for such a long period.

E.9 Health hazards

Potential, direct health risks

Operation of a P&T system, assumed it would work as advertised, would greatly increase the health risks posed by nuclear power for several reasons. Planned (authorised) discharges will increase. The chances of unplanned, but inevitable discharges and of occurrence of accidents involving large amounts of radioactivity will grow rapidly with time. Consequently the dispersion of radioactivity into the environment and its adverse health effects will increase.

- The total amount of radioactivity per unit useful energy from nuclear power delivered to society would be a multitude of the amounts generated by the current nuclear power plants.

- In the P&T cycle massive amounts of radioactive materials would remain very mobile for very long periods (decades to centuries). Chemically mobile because at any moment a substantial part of the radioactive materials is present in solution, and physically because all materials are continuously transported between the facilities of the P&T cycle.
- The chance of accidents and substantial releases of radioactive materials into the human environment increases drastically, compared to a situation without P&T, because more radioactive materials are stored in temporary storage facilities. These facilities would became ever more unsafe with time due to inevitable degrading processes of the materials and structures.
- In each cycle the gaseous fission products are discharged into the environment without retention. In addition a substantial part of the highly soluble fission products are discharged in the liquid effluents, together with appreciable part of the actinides. The population living in the vicinity of installations of a P&T system would be chronically exposed to ever increasing amounts of radioactivity.
- The total volume of the radioactive waste increases greatly in the P&T cycle. Each time the compact fuel and target elements are reprocessed (partitioned) the radioactive contents are distributed over volumes of liquids many times larger than the original volume. The radioactive solids (cladding hulls and insolubles) to be stored increase by every cycle.
- The amounts of dismantling wastes from all installations of the P&T cycle, highly contaminated with all kinds of radionuclides including actinides, are many times more than from the power plants themselves.
- In the P&T cycle substantial amounts of plutonium would be circulating, greatly enhancing the risk of nuclear terrorism.

Present, indirect health risks

As long as the nuclear industry promotes P&T systems as a solution to the radioactive waste problem, the attention of the public and politicians is diverted from the urgency of the safe disposal of the existing radioactive waste, to false expectations from concepts possible only in cyberspace. Every euro or dollar pumped into the development of a P&T system is wasted money and postpones the only possible safe solution of the waste problem: storage in a deep geologic repository.
Each week of postponement enhances the chances of accidents involving large amounts radioactive waste. Each week of postponement the unintended but unavoidable discharges of radioactive materials into the environment are continuing.

E.10 Summary

Technical features of partitioning & transmutation

The concept of partitioning and transmutation systems involves an extremely complex cycle of advanced unproved technical processes. At this moment the P&T cycle exists only in cyberspace.

In its publications to the public and politicians the nuclear industry seems to introduce the P&T concept as if its realisation is just a matter of technical development, money and some time, one or two decades. Technically the P&T cycle is similar to the ill-fated U-Pu breeder cycle, albeit much more demanding. In view of the experiences and investments of the U-Pu breeder cycle during the past 50 years, the optimism of the nuclear industry seems not to be based on any emprical evidence.

An inherent limitation of the P&T concept is that not all long-lived radionuclides can be transmuted into short-lived or stable nuclides for physical reasons.

Even with a perfectly operating transmutation system it would take centuries of continuous and flawless operation to reduce a given quantity of certain long-lived fission products and actinides to 1% of the original quantity.

In the most optimistic scenario it would take many decades to a century of continuous and flawless operation to reduce a given quantity of actinides to 1% of the original quantity. This would require a unproved type reactor as transmuter (Accelerator-Driven System), which is not suitable for transmutation of fission products.

It is not possible to completely eliminate the actinides from spent nuclear fuel for two reasons:
- In a perfectly operating P&T system an infinite number of recycles of target elements would be needed.
- Due to inevitable process losses in the partitioning plant and target fabrication plant a part of the actinides will be lost in the waste streams and into the environment.

During the last decade the focus of the international investigatings on P&T systems has shifted to advanced fast reactors as transmuter. Fast reactors are not suitable for transmutation of long-lived fission products, only for transmutation by fission of actinides. This observation might imply that the nuclear industry has abandoned the idea to transmute all long-lived radionuclides in spent fuel.

The concept of an advanced fast reactor with closed fuel cycle which has farthest progressed is a direct descendant of the Liquid Metal-cooled Fast Breeder reactor (LMFBR). The LMFBR cycle proved to be technically not feasible, and logically also economically not viable, after 4-5 decades intensive research in a number of countries and investments of hundreds of billions of dollars.

Radioactivity

The radioactivity of the materials from spent fuel, measured in Bq/kg, would strongly increase if the long-lived radionuclides are transmuted into short-lived ones for two reasons:
- A given amount of short-lived radionuclides emits more radiation than the same amount of long-lived radionuclides, because more decay events per second occur. The radioactivity of the original amount of short-lived radionuclides decreases faster due to the higher decay rate
- The number of radionuclides increases by activation reactions and by fission of the actinides.

P&T systems coupled to power reactors would drastically increase the amount of radioactivity generated per unit net energy delivered to the grid. In addition to the radionuclides in the fission products, actinides and activation products generated in the nuclear power reactors, the P&T system produces itself these categories of radionuclides in large amounts.

By application of P&T systems coupled to power reactors the volume and mass of radioactive waste would be multiplied. The radioactive contents of spent fuel and target elements would be dispersed over massive volumes and masses chemicals and materials.

By application of P&T systems coupled to power reactors the amounts of radioactive materials discharged routinely into the environment per unit net energy delivered to the grid would drastically increase.

By application of P&T systems coupled to power reactors the entropy of the radionuclides from spent fuel would be multiplied. The radionuclides are redistributed over large volumes and mixed with other, non-radioactive materials, and a part of the radionuclides is released into the environment. By these processes the entropy of the radionuclides increases immensely and consequently they become uncontrollable.

The P&T concept is proposed starting from the basic assumption that spent fuel is dangerous mainly because of the presence of actinides and long-lived radionuclides and that nuclear waste would not pose problems if the decay time of the radioactive contents could be reduced to several centuries.

The view of the nuclear industry that highly radioactive waste could safely be stored in above-ground facilities during a period of 400-1000 years, is not based on any evidence.

P&T and the Second Law

Analysis of a hypothetical P&T system operating according to design specifications proves that the concept as advertised apparently does not allow for the Second Law of thermodynamics, one of the most fundamental laws of nature.
From the Second Law follow, among other, several observations which are relevant for the functioning of a partitioning and transmutation system and of closed-cycle reactors:

- It is not possible to separate a mixture of different chemical species completely into 100% pure fractions.
- Fractionating a mixture becomes less efficient and takes more effort and energy as:
    the mixture contains more different species
    the chemical properties of the constituents are more alike
    the concentrations of the species in the mixture are lower
    the radioactivity of the mixture is higher.
- Fractionating a mixture is inevitably coupled to losses of the desired species of the fractions into the waste streams.
- It is not possible to produce materials which are 100% pure.
- It is not possible to produce materials the properties of which remain 100% predictable over prolonged periods.
- Every piece of equipment, machines and/or electronics, will fail after some time; it is not possible to predict at which moment they will fail.
- By means of extensive ‘preflight’ testing it is possible to grade up the reliability of components, but at the expense of dedicated effort and investment of energy, the more the higher the reliability requirements are.
- Due to spontaneous processes the quality of materials and euipment will always degrade with time (ageing). Upgrading the quality requires dedicated effort and investment of energy.

Cost

It is not possible to estimate the costs of a P&T system, if technically feasible at all. The fact that the dismantling of one reprocessing plant likely will cost at least a €100bn and that one P&T system has to replace its reprocessing (partitioning) plant several times during its operational lifetime, may give a first indication.

Risk assessment

In its promotional publications on P&T as solution to the nuclear waste problem, which anyway deny the nuclear waste to be a problem, the nuclear industry seems to ignore a number of features of a hypothetical P&T system which would have far-reaching safety consequences when a P&T system would come into operation. To name a few:

- In an operating P&T cycle massive amounts of radioactivity (tens of reactor core equivalents) are in highly mobile state for long periods: many decades to centuries.
- In an operating P&T cycle large amounts of highly fissionable nuclides, plutonium and higher actinides, are present in mobile state. This could enhance the chances of nuclear terrorism.
- The total amount of radioactivity in a nuclear power supply system with P&T cycle would be several times as large as in a system without P&T cycle (power reactors only).
- The radioactive materials from power reactors and P&T system would be distributed over vast amounts of liquids and solids: the total volume of the radioactive waste would be multiplied.
- Gaseous and highly soluble fission products would be discharged into the environment at the partitioning plant (routine discharges), not only from the spent fuel from the power reactors, but also from the transmuter reactors. The standards of authorised discharges are actually designed by the nuclear industry itself.
- During the operational lifetime of a hypothetical P&T system all installations of the cycle, the partitioning plant, the fuel and target element fabrication plant and the transmuter reactor, have to be replaced by new ones repeatedly. Every technical installation has a limited operational life and a P&T system would have a long operational lifetime. During the decommissioning and dismantling of these installations massive amounts (hundreds of thousands of tonnes) of highly radioactive waste would be released. These inevitable consequences of the introduction of a P&T system would imply considersble safety and health risks, costs and energy investments.
- Apparently the storage of highly radioactive waste during a period of 400-1000 years in above-ground facilities would not pose a problem, in the view of the nuclear industry. It is not clear how the nuclear industry intends to achieve a safe storage of massive amounts of highly radioactive materials during a period of that length. Experiences regarding storage of radioactive waste during the past 60 years are far from encouraging.
- After removal of the long-lived radionuclides from the nuclear wastes by P&T and a storage period of 400-1000 years the wastes would be as radiotoxic as uranium ore. Apparently the nuclear industry assumes this to be a safe level. It is not clear how the radiotoxicity of uranium ore has been calculated, nor how the radiotoxicity is defined and calculated of the nuclear wastes resulting from a P&T cycle after a storage period of 400-1000 years.

Evidently above mentioned features would greatly increase the chances for the public of exposure to radiation and contamination with all kinds of radionuclides, including chances of large accidents, and consequently would greatly inrease the health risks of nuclear power.

Energy balance

The P&T cycle comprises two very energy-intensive porocesses: partitioning and fuel and target fabrication. In addition all facilities are to be dismantled after their operational lifetime. This may be needed several times during one sequence of cycles (one or more centuries) of one P&T

system. Decommissioning and dismantling likely will require very large energy investments. It is not certain if the intended transmuter reactors will deliver net energy. Most likely the P&T system will be an energy sink to such extent that a nuclear energy system consisting of a number of power reactors and associated P&T systems will be a sizeable energy sink instead of a net energy source.

Human resources

A P&T system has to operate for at least several centuries to treat the waste from a given reactor after closedown of that reactor. Great numbers of highly skilled people will be needed to perform these demanding processes. Even if the last nuclear power plant would be shut down today, the economy has to sustain a nuclear workforce for centuries to come. This workforce will not contribute to any improvement of the energy supply. Its sole task is to prevent the nuclear legacy from going disastrous. One may wonder if enough young people would opt for the required rigorous education and training and if a free market-oriented economy would be able to support such a workforce for such a long period.

E.11 Conclusions

1
By implementation of P&T the long-lived component of the radioactivity of spent fuel would be transformed into short-lived. This would result in a sharp increase of the total amount of radioactivity in nuclear waste during the next centuries and less radioactivity after that period. It is not clear on which grounds the nuclear industry bases its view that this situation would be safer and would pose less health hazards than disposing off spent nuclear fuel in deep geologic repositories, packed in durable containers, as soon as possible after removal from the reactor and without reprocessing.

2
Health hazards would increase by the introduction of P&T systems, compared to the hazards caused by a nuclear energy supply system without P&T, due to:

- generation of a larger amount of radioactivity per kWh electricity delivered to the consumer
- distribution of the human-made radioactivity over larger volumes and masses of materials
- more radioactivity in chemical and physical mobile state during prolonged periods
- more discharges of human-made radioactivity into the environment
- more risks by human behaviour.

3
The introduction of P&T systems would introduce higher risks of proliferation and nuclear terrorism, due the larger amounts of plutonium circulating between the facilities of the P&T cycle, during extended periods.

4
Each of the proposed partition & transmutation concepts, if they were to operate as advertised, should comply with the following requirements, each of which may considered to be a conditio sine qua non:

- perfect separation of the constituents of spent fuel into pure fractions (partitioning),
- fine-tuned operation of the three components of the P&T cycle – partitioning, fuel + target fabrication and transmuter reactor – together,
- flawless and continuous operation of the whole cycle during many decades to centuries.

None of these conditions can be met due to fundamental limitations following from the Second Law of thermodynamics.

5

Apart from the fundamental limitations, a P&T system exhibits some features, each of which might easily be seen as prohibitive:

- a negative energy balance
- extemely high and uncontrollable costs
- creation of a long-term commitment to very demanding and costly activities, meaning a heavy economic burden for future generations.

The question rises for what reasons the nuclear industry promotes this concept. Fund raising?

as an excuse (“new technology has to be developed”) to postpone investments in the really possible solutions of the radioactive waste problem?

Does the nuclear industry not see the large hurdles preventing the realisation of an operating P&T system?

would take centuries to reduce actinides to 1% (ideally), so what would be the benefit of P&T?

This observation may imply that the nuclear industry has abandoned the idea to transmute all long-lived radionuclides in spent fuel.
What reason for p&t?

The principal purpose of a P&T system is said to be the reduction of the hazards posed by spent nuclear fuel. In practice, assumed a P&T cycle would operate according to the design specificationshe reverse turns out to be true.

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ANNEX F

Thermodynamics, entropy and Second Law

F.1 Thermodynamics

Energy plays a key role in our universe:
- All changes in the observable universe are accompanied by energy conversions.
- All energy conversions are governed by the thermodynamic laws.

Thermodynamics is the science of energy conversions and is at the base of all sciences.

From the observations above follows the conclusion:

> *All human activities are accompanied by energy conversions and consequently all human activities are governed by the laws of thermodynamics.*

This Annex introduces the some basic notions and laws of thermodynamics:
- system
- energy and the First Law
- entropy
- Second Law

In the context of this study the Second Law of thermodynamics is important.
It should be noted that some of the definitions used in this study are related to the applications in this study and may look somewhat different from the definitions applied in publications with a more mathematical approach, addressing highly specialized subjects involving thermodynamics.

System

In thermodynamics a system is defined as the observed quantity of matter and space which is of interest in the context of a given scientific study. The system plus its surroundings is sometimes called the universe. If we want, for example, to analyse the energy balance of a car, then the car is the system and the rest of the world is called the surroundings.

It should be noted that the word ‘system’ is also used with other than thermodynamic meanings: in figurative senses such as the system in a language, and in technical senses, for example the operating system of a computer.

System boundaries

Any study involving a system in whatever sense has to meticulously define the boundaries of its system, not only in spatial and material sense but also with regard to its time horizon. Observations and conclusions which may be valid in one given system, need not to be valid in another system with different boundaries. Many scientific studies utilize some kind of model: a simplified description of the observed system. Such a scientific model is based on a number of assumptions and choices made by the investigator. The validity of a model is strongly related to the boundaries of the system it is conceived for.

F.2 Energy and the First Law

Energy

Energy is a conserved quantity and a basic notion: its definition cannot be deduced from other notions. A usable description of energy is:

Energy is the entity making changes possible.

Energy can manifest itelf in various forms: electricity, kinetic energy, heat, radiation and chemical energy. Potential energy is the energy, not readily observable, present in fossil fuels and uranium, but also in a given mass at a certain height above the surface of the earth. The energy in fossil fuels is actually chemical energy, which is usually set free as the combustion heat of the fuels.

The SI unit of energy, used throughout this study, is the joule, symbol J. Other energy units are ambiguous, because of different possible definitions. For example, there exist six definitions of the British thermal unit (Btu) and five of the calorie (cal). A joule is a small unit (about the energy of one heart beat), so usually large multiples are used.

First Law

The First Law is the well-known law of energy conservation:

Energy cannot come into being from nothing, neither can be destroyed.

The energy of the observable universe, in a thermodynamic sense, is constant.
Energy can be transformed from one form into another, for example light into electricity by a PV solar cell and heat into mechanical energy by a car engine, but energy can never been lost. Confusion may be evoked by expressions in daily life, like ‘energy generation’ and ‘energy loss’. These terms are related to the quality of energy, its usefulness.

F.3 Entropy

Spontaneous changes

Some changes happen spontaneously, other don’t. A cup with hot tea cools down to the temperature of the surrounding air; a cup of cold tea never gets hotter, cooling down the surrounding air. A piece of charcoal (carbon) burns to hot carbon dioxide, but an amount of hot carbon dioxide never forms spontaneously a piece of pure carbon. Reheating the tea or reconversion of the carbon dioxide into carbon is possible only by doing work: dedicated effort and useful energy.

Not all spontaneous processes will start spontaneously: sometimes a small amount of activation energy is needed to get the process going. For example one spark is needed to ignite any amount of oil, wether 1 gram or millions of tonnes: once started the spontaneous process continues until the last drop of oil has burned.

When a change occurs, the total energy in the universe remains constant, according to the First Law. Spontaneous changes are always accompanied by a reduction of the quality of the involved amount of energy: during the change the energy quality is degraded to a more dispersed form. Spontaneous processes are consequences of the natural tendency of the universe towards

greater entropy, a greater dispersion (randomness) of matter and energy.
The reversal of a spontaneous process, if possible, would require dedicated effort and useful energy.

Definition of entropy

Entropy is a vital notion in thermodynamics. Entropy is based on a probabilistic principle at the level of atoms and molecules, expressed in the famous formula of Boltzmann:

$$S = k \ln W$$

here is: S = entropy of a given system
k = constant of Boltzmann = 1.3807 $J.K^{-1}$
W = number of microstates of the system

This formula will not be used explicitely in this study, so above quantities will not be explained here.
The probabilistic character of entropy is based on the quantummechanical concept of quantisation of matter and energy: mass flows and energy flows occur in quanta, smallest units, for example atoms and light quanta.
The probabilistic aspects of entropy are of paramount importance in chemistry, biology and environmental sciences. For that reason the following description of entropy, based on the formula of Boltzmann, may be useful:

> *Entropy is a measure of the dispersion and randomness of matter, of energy and of oriented flows within an observed system. A higher grade of dispersion means a higher entropy.*

Entropy changes

Entropy is a measurable quantity, in principle, but the absolute value of the entropy of a given system is not required to be known in the decription and analysis of processes. Entropy itself is not directly observable, we can only notice entropy *changes*. In human actions and in the economic system only changes are involved, so of interest are just energy *changes* and entropy *changes*.
What is here called an entropy effect is actually an entropy *change* of the observed system. An entropy increase of the system means that the mass particles and energy quanta of the system get more randomly dispersed. An entropy decrease implies a more ordered state of the mass particles and energy quanta.

Entropy changes in large and complex systems are difficult to quantify. Fortunately, for getting insight into the global environmental issues, including the climate change, even entropy changes need not to be quantified: just qualitative or semi-quantitative considerations on entropy changes are generally appropiate. To understand the consequences for the biosphere and human environment we only need to know if a given natural process or human action will result in an entropy decrease or increase of the observed system.

Entropy and chance

The probabilistic principle behind the notion *entropy change* can be illustrated at the level of molecules and atoms. An entropy increase of a system means that the state of the particles of the system becomes more probable in the probabilistic sense than the state before. A state of the molecules and atoms of the system that is more set by chance means less ordering, more

dispersal of matter and energy and less quality of the system. Less quality of a system means less functionality of that system to man. A drop of perfume in a room has a lower entropy and higher functionality than the same amount evaporated in the same room.
On the other hand, a decrease of the entropy of a system means a transition to less probable state of the particles in the system, less set by chance, and consequently a higher grade of order and a higher functionality.

Energy can also become dispersed and consequently loose quality and functionality. For example, the conversion of electricity into heat by resistance of the conducting wire, is an entropy effect. The electrons in the wire flow in one direction with a certain force. By interactions with the atoms of the wire the directed movement of the electrons is converted into random movements of the atoms: heat. The heat is randomly radiated into the surroundings of the wire. The energy in the electric current is converted into heat which has less functionality: it cannot do work anymore, for instance to power a computer.

F.4 Entropy and functionality

In many cases entropy may be perceived as a measure of chaos: higher entropy means more chaos and more mess. With regard to human activities an increase of entropy may also be taken as a loss of quality and usefulness. For that reason entropy may be described in non-physical terms as a measure of '*mess and uselessness*'.
When a steel tube rusts and decays into a pile of brown grains, the mess and uselessness of the original amount of steel have increased, or in thermodynamic terms: the entropy of the system has increased. The amount of iron in the observed system has not changed: the iron atoms of the original tube are still present in the pile of rust grains.

Materials in the context of economy and society generally are processed materials, such as metals, chemicals and plastics. These materials have desired and predictable properties, a high functionality and usefullness. We need pure drinking water, food without contamination, medicines with very predictable effects, computers, cars and airplanes that will function reliably for long times, a clean environment.

Usefullness has to do with quality, functionality and predictability of the behavior of substances and constructions. The higher quality, the better a material satisfies specific needs, is more durable and has less chances of failures. A positive entropy change in a system, that is an increase of the entropy of the system, means a degradation of the functionality of the system.

A computer chip with a specific function that has to operate for many years exactly conform the specifications, need to be made from extremely pure materials and these materials are to be assembled in a highly specific and stable configuration. The atoms in the system (the chip) need to be in an exceedingly ordered and specific configuration, which is highly improbable from a stochastic viewpoint. When the ingredients of a computer chip would be mixed in a reaction vessel at an appopriate temperature, one can be sure that no chip will be found in the vessel afterwards.

On atomic level a higher quality of a material (system) means that the order is more specialised and less probable from a stochastic viewpoint. Consequently a higher quality corresponds with a lower entropy of the system. Upgrading an amount of raw material (material as found in nature) into a useful substance or a piece of equipment implies lowering the entropy of that amount of raw material.

F.5 Second Law

Stochastic processes

At the base of the Second Law lies the notion that any spontaneous process is a stochastic process, that will result in a more randomized distribution of particles and energy quanta of the system than before. A stochastic process occurs at random, without any form of external influence, and is fully determined by chance.
The following phrase is a nice metaphor of entropy and a spontaneos process:

> *Any fool can pour a cup of tea into the ocean, but a thousand wise men cannot pull it out again.*

Examples of spontaneous processes are: the dispersion of CO_2 from burning fuel into the atmosphere, the rusting of steel in the open air and the decay of dead organisms.

Second Law

Entropy and the Second Law are playing a crucial role in the events and phenomena in our visible world. To understand the effect of the Second Law in the context of this study a full comprehension of the notion entropy is not necessary, for it will be used only qualitatively or semi-quantitatively.
Up until now no phenomena have been observed within the observable universe which would violate the Second Law, For that reason the Second Law is accepted in the natural sciences to be valid within the observable universe.

The Second Law can be formulated in different ways. Evidently all correct formulations are based on the same principle: the dispersion of matter and the randomizing of oriented energy flows by any spontaneous process. A basic formulation of the Second Law is:

> *With every change the entropy of the universe increases.*

In respect of processes of human scale the following formulation is useful:

> *In a system without input of energy and matter from the outside any spontaneous process will increase the entropy of the system, and so will decrease its quality and usefulness.*

From above definition follows a consequence of the Second Law which is important for environmental studies:

> *Any spontaneous process within the biosphere without input of energy and materials from the surroundings (outer space) increases the entropy of the biosphere.*

Two approaches

Entropy and the Second Law are not very popular in the world of science and technology. One reason may be the fact that many observed phenomena in nature and technology can be explained without these two concepts. A second reason might stem from the way entropy and the Second Law are introduced: there is a mathematical approach and a statistical/probabilistic approach.

The mathematical method is based on mathematical definitions of entropy and Second Law and applies only to cases under very specific conditions in simple and surveyable systems. The mathematical method applies entropy and Second Law quantitatively. However, the mathematical method is not easy to access, even for scientists who are not specialized in the

field.
The probabilistic method is based on the phenomenological essence of entropy and is often used in chemistry and biochemistry. The probabilistic method is suitable for qualitative and semi-quantitative application of entropy and Second Law in assessments of complex systems, such as (bio)chemical systems, ecosystems and the biosphere as a whole. This method is better accessible than the mathematical method.
This study follows the course of the probabilistic approach of entropy and the Second Law.

Importance of the Second Law

The notions entropy and Second Law have little relationships with daily life, although their effects are common. We are used to see iron rusting, water evaporating and a dead organism decaying. In daily life we do not need elusive notions such as entropy and the Second Law to explain the observed phenomena. In many fields of natural sciences the same observation holds true: a lot of phenomena in nature can be described and explained based on familiar natural laws without involving entropy and the Second Law. However, entropy and the Second Law are essential in explaining the basic processes in chemistry, biochemistry and physics.

The reason why the notions entropy and the Second Law are now playing a prominent part in environmental sciences is the scale of the human activities in the biosphere at this moment. Human behaviour has observable global effects. The magnitude of the human activities in relationship with the size of biosphere, as a thermodynamic system, forces us to go to the basics of science. Human activities are not negligible anymore in comparison with the natural processes in the biosphere and are in fact competing with them.

F.6 Consequences of the Second Law

Ageing of materials and structures

An example how the Second Law works out is what happens to a steel structure if left unattended in the open air for a long period. The structure will become useless, due to the spontaneously occurring deterioration of the steel and decay into a pile of rust. The entropy of the system (i.c.the steel structure) will spontaneously increase, according to the Second Law. Increasing entropy means decreasing reliability.

Maintenance is required to prevent the structure from becoming useless; in fact maintenance can only delay the moment a given structure has to be replaced. Ageing processes are unavoidable, also under conditions of good maintenance.
Even when starting from perfect materials, which is not possible as explained below, ageing will occur due to mechanisms such as:

- Diffusion of foreign atoms into the material, for example tritium atoms diffusing into the zirconium cladding of the fuel elements and into the steel of the nuclear reactor vessel.
- Nuclear radiation: atoms are displaced by energetic radiation and consequently disturb the most stable crystal structure of metals; neutron radiation has a more drastic effect, because by neutron capture reactions atoms of other elements come into being, that may be not compatible with the original material.
- Displacement of atoms of the material under thermal and/or mechanical stress. Evidently this effect disturbs the optimal crystal structure of the material and consequently affects its properties negatively.

Figure F1
Consequences of the Second Law. This photograph of a steel tube on the sea shore shows what irrevocably will happen to any man-made structure. Left unattended any structure will spontaneously degenerate by corrosion, weathering and wear, and in the long run human-built structures will decay into useless dust.

Separation and purification processes

An important consequence of the Second Law with respect to separation and purification processes is:

> *Separation of a mixture of different chemical species never goes to completion. Consequently it is not possible to separate a mixture into its pure constituents without losses. The amount of useful energy required for separation increases with the number of chemical species in the mixture and with the desired purity of the separated constituents.*

From this observation follows that 100% pure materials are not possible, there always be some contaminants left. At the expense of large dedicated efforts and the investment of much useful energy, very pure materials can be produced, for example silicon chips and medicines. Purity of materials is an economic notion: which purity grade is affordable?

Mineral energy sources

Another consequence of the Second Law, important in the context of the generation of useful energy from mineral energy sources (fossil fuels and uranium) is:

> *The generation of an amount of useful energy from a mineral energy resource (fossil fuels, uranium) inextricably generates more disorder and more loss of quality (= more entropy) of the biosphere than can be compensated for by the produced amount of useful energy.*

As explained above, the burning of fosssil fuels and the fission of uranium are spontaneous processes, once started, without input of materials and energy from outside the biosphere. As a result the entropy of the biosphere, the system in which these spontaneous processes occur, increases and increasing entropy means more environmental damage.

F.7 Observable entropy effects in the biosphere

Increase of the entropy of the biosphere caused by human activities manifests itself as a increasing deterioration of the environment and loss of quality of ecosystem services. In fact, all anthropogenic environmental problems are entropy effects. This is not difficult to recognize, for they are caused by dispersion of matter and energy and by consumption of the usefullness of ecosystem services. Under the present conditions human activities are degrading the quality of ecosystems, causing loss of functionality.

Examples of anthropogenic entropy effects are:
- dispersion of CO_2 and other human-made greenhouse gases throughout the atmosphere
- pollution of air by dust, soot and acidifying gases
- pollution of ground water, rivers, lakes, sea, air and soil by anthropogenic chemicals
- oil spills
- dispersion af radioactive materials into the air, water and soil
- erosion of arable land, loss of topsoil, degradation and decline in soil fertility
- washout of phosphate fertilizers into rivers and sea
- desert forming by overgrazing of grasslands
- decline of biodiversity
- decline of fish populations in the sea
- destruction of ecosystems by mining activities
- deforestation
- loss of irreplaceable materials,such as platinum and phosphates
- rising global temperatures
- fresh water shortages

Most of these entropy effects are irreversible on human timescales.

The Second Law provides us with a solidstandard in the quest for sustainable energy and sustainable economic system.

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ANNEX G

Energy cliff and thermodynamic scarcity

G.1 Energy cliff

Dilution factor

Metals and other minerals are recovered from ores in the earth's crust. The extraction of a metal from its ore consumes more useful energy and ordered materials (chemicals, equipment, etcetera) as the ore grade is lower. Evidently the amount of rock to be mined, milled and chemically treated to recover 1 kg of a given metal increases with lower grades: to recover 1 kg metal from ore with a grade of 1 kg metal per Mg (1 megagram = 1 metric tonne) rock the amount of processed rock is ten times greater than when extracted from ore at 10 kg metal per Mg rock.

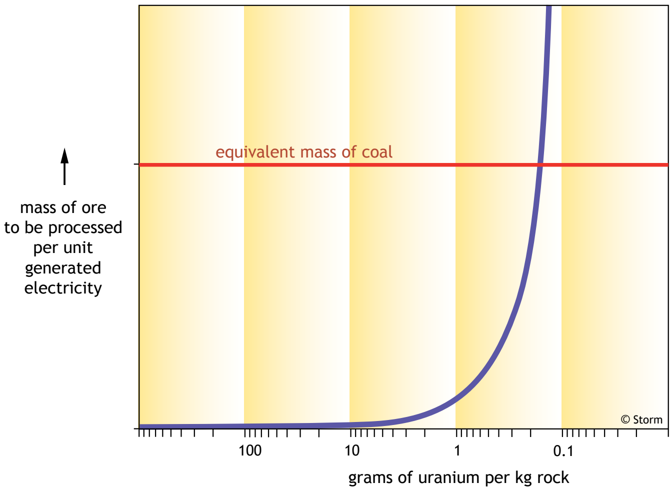

Figure G1
The dilution factor of the recovery of uranium from ores. The horizontal axis has a logarithmic scale, the vertical axis has a linear scale. At grades of about 0.2 grams of uranium per kilogram rock, as much uranium ore has to be mined, milled and chemivally processed to deliver as the amount of coal to be mined and burned to generate the same quantity of electricity: the coal equivalence.

Recovery yield

This dilution effect is exacerbated by a second effect: the extraction yield decreases with decreasing ore grade. The fraction of the metal that can actually be extracted from its ore goes down with falling ore grades (for details see Q9 p ## and Part ## of Q6). The decrease of the extraction yield with decreasing metal content in ores is a consequence of the decreasing separation efficiency at lower grades. This phenomenon follows from fundamental laws of nature, especially the Second Law of thermodynamics (see Annex ###).

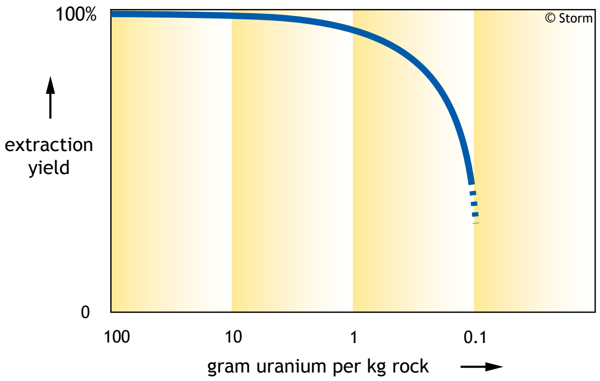

Figure G2

The recovery yield, or extracrion yield, of the recovery of uranium from ores as function of the ore grade.

Energy cliff of uranium

Uranium, also a metal, is almost exclusively used as energy source. The amount of useful energy which can be generated from 1 kg uranium leaving the mine has a fixed value within small margins. Above observations (dilution effect and extraction yield) imply that the amount of energy consumed for recovery of 1 kg uranium is larger, the lower the grade of the uranium ore from which that kg is recovered. At a certain low grade the energy consumed equals the potential energy production. This phenomenon is called the *energy cliff*. The critical ore grade lies at 100-200 g uranium per Mg rock.

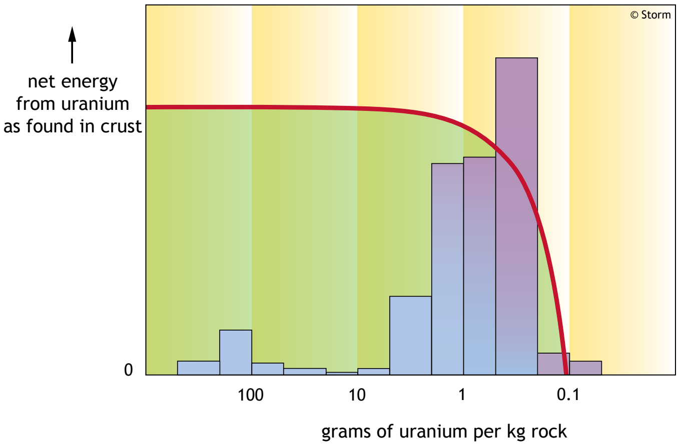

Figure G3

The energy cliff of uranium resources. Below a given ore grade no net energy can be extracted from a uranium deposit as found in nature.

Fossil fuels

A similar phenomenon as the energy cliff of uranium occurs with fossil fuels as well. For example the recovery of oil from tar sands or deep wells in the Arctic and of gas from shales (fracking) consumes an appreciable fraction of the energy content of the recovered fuels. The same holds true for coal, the grades of the available coal resources decrease with time – from anthracite to bituminous to brown coal – so more coal has to be mined to get the same amount of energy (heat). The fraction of the energy content needed for recovery and refining of fossil fuels increases with time for the same reasons as pointed out above in case of uranium resources. In his publication Jeff Rubin [Q503] discussed this development from an economic point of view.

G.2 Thermodynamic scarcity of energy and materials

Quantitatively metal resources cannot run out completely: there will always some amounts of a given metal be left in the crust. However, metals are becoming scarcer in another sense. The easiest exploitable metal ore resources are always exploited first, because these offer the highest return on investments. New discoveries of ore deposits will seldom be of the same quality – high ore grade, large size, shallow depth beneath the surface, easy accessible, favourable mineralogical properties – of the resources already exploited, generally the new discoveries have a lower quality. As a result the resources left over are harder to exploit. This implies that the amount of energy and ordered materials consumed per kg recovered metal increases with time.
This effect is reinforced by the observation that the generation of one unit of useful energy itself is requiring more ordered materials and useful energy with time.

When the investments of energy and ordered materials to recover 1 kg of a given metal Me become higher, a greater fraction of the energy flows in the world economic system are needed for extraction of that kg metal Me from the crust. This is an exponential growth, which goes still steeper when the consumption of that metal Me rises in a growing economy.

Obviously higher specific consumption of energy and materials, per kg obtained mineral, will cause a rise in the financial production cost of minerals and so of the materials produced from that minerals. History shows there is no simple relationship between energy intensity and market prices of minerals and materials, but generally and in the long run more effort per kg product translates into higher prices.

Materials will become scarcer in a qualitative sense: the energy intensity may grow to a level prompting the question: how much energy is affordable in the economic system to use a certain mineral for a given purpose? Eventually the availability of useful energy is the limiting factor for economic activities. A growing part of the useful energy generated in the world economic system is needed to produce useful energy from its mineral sources, as pointed out above, so a shrinking part will be left for other purposes.

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ANNEX **H**

IAEA, ICRP and UNSCEAR

H.1 IAEA

The International Atomic Energy Agency (IAEA) is an international organisation that seeks to promote the peaceful use of nuclear energy, and to inhibit its use for any military purpose, including nuclear weapons. The IAEA was established as an autonomous organization on 29 July 1957. Though established independently of the United Nations through its own international treaty, the IAEA Statute (http://www.iaea.org/About/statute.html), the IAEA reports to both the UN General Assembly and Security Council. Eighteen ratifications were required to bring the IAEA's Statute into force on 29 July 1957.
Total Membership: 159 (as of February 2013). The Democratic People's Republic of Korea (DPRK), which joined the IAEA in 1974, withdrew its membership of the IAEA in 1994. (http://www.iaea.org/About/Policy/MemberStates/)

The IAEA Mission Statement of the International Atomic Energy Agency reads:

* is an independent intergovernmental, science and technology-based organization, in the United Nations family, that serves as the global focal point for nuclear cooperation;

* assists its Member States, in the context of social and economic goals, in planning for and using nuclear science and technology for various peaceful purposes, including the generation of electricity, and facilitates the transfer of such technology and knowledge in a sustainable manner to developing Member States;

* develops nuclear safety standards and, based on these standards, promotes the achievement and maintenance of high levels of safety in applications of nuclear energy, as well as the protection of human health and the environment against ionizing radiation;

* verifies through its inspection system that States comply with their commitments, under the Non-Proliferation Treaty and other non-proliferation agreements, to use nuclear material and facilities only for peaceful purposes.

Official publications of the IAEA have to be approved by all member states of the IAEA, consequently the IAEA canot be considered an independent scientific institute.

H.2 ICRP

The International Commission on Radiological Protection (ICRP) is an advisory body providing recommendations and guidance on radiation protection. It was founded by the International Society of Radiology (ISR) in 1928 and was then called the International X-ray and Radium Protection Committee (IXRPC). It was restructured and given its present name in 1950 (ICRP 109 2009 Q543). The ICRP has more than 200 volunteer members from about 30 countries. According to the ICRP publication *1959 Decisions* (www.icrp.org):

> 'The Commission has an official relationship with the World Health Organization and the International Atomic Energy Agency. There has been close co-operation with the United Nations Scientific Committee on the Effects of Atomic Radiation, ...'

International System of Radiological Protection

The International System of Radiological Protection, developed by the ICRP (www.icrp.org) is based on:

- current understanding of the science of radiation exposures and effects, and
- value judgments. These value judgments take into account societal expectations, ethics, and experience gained in application of the system.

The system of radiological protection that is used across Europe and worldwide is based on the recommendations of the ICRP and the International Commission on Radiation Units and Measurements (ICRU), according to SCENIHR 2012 Q533. These recommendations are based on three fundamental principles:

- justification
- optimisation and
- dose limitation.

> “The principle of justification requires that any decision that alters the radiation exposure situation should do more good than harm; in other words, the introduction of a radiation source should result in sufficient individual or societal benefit to offset the detriment it causes.”
>
> “The principle of optimisation requires that the likelihood of incurring exposures, the number of people exposed and the magnitude of their individual exposure should all be kept as low as reasonably achievable, taking into account economic and societal factors. In addition, as part of the optimisation procedure, the ICRP recommends that there should be restriction on the doses to individuals from a particular source and this leads to the concept of dose constraints.”
>
> The principle of dose limitation requires that
> “the dose to individuals from planned exposure situations, other than medical exposure of patients, should not exceed the appropiate limits recommended by the Commission.”

As part of the system of radiological protection ICRP 103 2007 Q544 defines three categories of exposure situations, namely:

- Planned exposure situations, which are situations involving the planned introduction and operation of sources. (This type of exposure situation includes situations that were previously categorised as practices).
- Emergency exposure situations, which are unexpected situations such as those that may occur during the operation of a planned situation, or from a malicious act, requiring urgent attention.
- Existing exposure situations, which are exposure situations that already exist when a decision on control has to be taken, such as those caused by natural background radiation.

The principles of justification and optimisation apply universally to all three exposure categories, whereas dose limits apply only to planned exposure situations, except some medical exposure situations. The ICRP recognises three categories of exposed individuals, workers, patients and members of the public. These categories of exposure are known as occupational, public and medical exposure.
It is not clear how the situations in the contaminated areas around Chernobyl and Fukushima would be categorized and what consequences such a categorisation would have for the population in the affected areas.

Scope of recommendations

Originally the exposure recommendations of the ICRP were designed for application in known radiation exposure situations: the planned introduction of radiation sources (X-ray and natural radioactive material, such as radium) for medical, scientific and technical purposes. These issues still are by far the main field of the ICRP, as may follow from the complete list of ICRP publications. Only a small number of the 140+ ICRP publications are dealing with exposures as a result of human-made radioactivity from nuclear power.

The main task of the ICRP seems to be the formulation of a legal framework for authorities and politicians how to cope with liabilities which may arise by exposure of people to radiation and/or radioactive materials (see for example ICRP 103 2007 Q544 and ICRP 111 2009 Q535).

Location and content of medical and industrial radiation generating sources are known and usually contain one specific radionuclide, and consequently these sources are fairly well under control. With the advent of nuclear reactors the exposure recommendations had to be adapted to situations with this new kind of radiation sources. In nuclear reactors dozens of different radionuclides are generated in the fission process and the content of radioactivity of one reactor is a trillionfold larger than the radiation potential of the sources before the nuclear era. During the past 60 years an amount of human-made radioactivity more than 10 million nuclear bomb equivalents has built up (section 2.2), dispersed over hundreds to thousands facilities and locations worldwide, an amount growing by some 300000 bomb equivalents each year. Consequently the scope of the radiological protection has grown immensely during the past 60 years and is still growing.

Uncontrollability of radiation sources

Chapters 3 and 4 addressed the mechanisms of dispersion of human-made radioactive material into the human environment. As every engineer knows: no technical installation is leakfree, leaks are unavoidable. The amounts of radioactive materials leaking into the biosphere are growing year by year for several reasons:

- increasing global inventory of human-made radioactiviy
- increasing number of storage locations
- unavoidable ageing of materials, accelerated by the nuclear radiation
- inadequate operational procedures and maintenace.

In addition to these unplanned releases radioactive materials are discharged into the human environment by:

- practice of authorised routine releases
- large accidents.

Concomittant problems are that routine releases of nuclear power plants (Chapter 3) are rarely measured and that unplanned discharges often are discovered only after a length of time, if at all.

The chance of exposure to radioactive substances and ionising radiation from human-made sources is approaching unit: on the Northern hemisphere people can be quite sure to be exposed to human-made radioactivity. The question is not if: the questions is: how much radioactivity and which radionuclides do we contract, and how could exposure be minimised or preferably prevented?

Protection

What does mean ‘radiological protection’ to the people involuntarily living in permanently contaminated areas? These areas are present not only in the vicinity of Chernobyl and Fukushima, but also near operating nuclear power plants and other nuclear facilities containing and releasing large amount of radioactive materials, such as reprocessing plants.

How is the perception by the general public of the word ‘protection’ when, after a nuclear incident, the allowed content of radioactivity in food and water is instantly raised, without scientific arguments, by a factor of 10, 100 or even more?

Releases of radioactive materials into the human environment by are often not measured, at leastnot during prolonged periods. Contamination of fish and food and drinking water with various kinds of radionuclides are sparsely monitored, and if so only for easily detectable radionuclides, such as cesium-137. The presence of tritium and carbon-14, two biochemically very active radionuclides, can be found only by special equipment. Not measured is not known.

H.3 UNSCEAR

See also UNSCEAR 2013b Q573

The United Nations Scientific Committee on the Effects of Atomic Radiation has been established 3 December 1955. The United Nations General Assembly has designated 27 States as members of the Scientific Committee. The mandate reads (UNSCEAR 2010 Report, Q531):

> “to undertake broad assessments of the sources of ionizing radiation and its effects on human health and the environment.”

At the time of the establishment of UNSCEAR, the only publicly known sources of atomic radiation were atomic bomb explosions, especially those of Hiroshima and Nagasaki, and from natural sources (uranium- and thorium-bearing minerals). No nuclear power plants were existing and the small number of military reactors in only a few countries of the world were hidden from the view of the general public. Note that the adjective ‘atomic’ has long been replaced by ‘nuclear’ in official publications, likely to avoid association with atomic bombs; just in the names of some international insitutes, such as IAEA and UNSCEAR, the adjective has not changed.

1 publication non-targeted and delayed effects
2 Chernobyl

3 From the text of UNSCEAR 2010 Report the conclusion [dringt zich op] that no ability is existing ‘to attribute scientifically risk and effects at both high and low doses, both for populations and for individuals’.

The Committee recognizes:

> ‘that its assessments of radiation exposures from electricity generation, while up to date and detailed for the nuclear fuel cycle, were out of date for the enhanced levels on naturally occurring radioactive material associated with the use of fossil fuels, and moreover had never been assessed in a comparable way for renewable energy sources.’

4 As pointed out in sections ### much unknown, may processes still not existing. Above statement of UNSCEAR that

5 one nuclear power station multiplies natural radioacvity billionfold (section ###), each year. How much uranium ore natural radioactivity mobilised? per kWh
how much by coal?
how much radioactivity involved in wind turbines, solar PV cells, only during fabrication, not during energy production ### 5. A nuclear power plant generates each year human-made radioactivity
6 how fair is ‘fair’?

Missing:
In the text of UNSCEAR 2010 [Q531] virtually no mention is made of human-made radioactivity: radiation sources generated by the fission process in nuclear reactors, only sources of natural

radiation and radiation from the atomic bombs in Japan are mentioned. (7)
Also missing in text are:

- references to nuclear power stations, let alone as sources of radiation exposures.
- large nuclear accidents, e.g. Chernobyl
- routine emissions npp's and mining
- releases of (human-made) radioactive materials into the environment from deteriorating waste storage facilites and leaking pipes and storage tanks.

What about the dumping of complete ship reactors and other radioactive waste into the sea?

8 The work of UNSCEAR seems to be focused on exposure to external radiation sources. However the Committee decided to focus work on internal emitters on tritium an uranium. This may be significant because ## tritium releases by npps, U by mining and use of depleted `u ###

9 Notably, the UNSCEAR considers 'assessments of radiation effects and risks especially for childern, and of the epidemiology of exposures of the public to natural and artificial environmental sources at low doses and low dose rates'.
This statement may refer, among other, to the results of the KiKK and GeoCap studies.

11 Many difficulties are encountered in attributing specific cases of disease to low-dose radiation exposure including:

- The lack of specificity in the type or characteristics of disease induced by radiation exposure
- The long delay (years or decades) between exposure and disease presentation
- The high spontaneous incidence of diseases associated with radiation in the ageing general population.

14 The Committee has judged that

> "the single most informative set of data on whole-body radiation exposure comes from studies of the survivors of the atomic bombings in Japan in 1945. The atomic bombing exposures were predominantly high-dose-rate gamma radiation with a small contribution of neutrons."

These studies started about five years after the bombings, so the deaths during these first five years are not counted. Would the methodology of these studies comply with the present scientific views and insights?
A second point is that these data sets are concerning only exposure to gamma rays and a small contribution of neutrons from an external source, and consequently do not comprise data on exposure to alpha and beta rays from radionuclides inside the body after contamination with radioactive materials via air, water and food.

Above statement of UNSCEAR implies, strikingly, that in the more than 60 years of nuclear activities, such test explosions of nuclear weapons, operation of military reactors and implementation of large-scale civil nuclear power, no better data sets have been generated. During the period following the studies of the survivors of the atomic bombings in Japan, the amount of human-ade radioactivity rose to the equivalent of more than 10 million Hiroshima bombs and is still rising at a present rate of about 300000 nuclear bomb equivalents a year.

15 The Committee judges that radiation-induced processes that can create long-lasting and transmissible effects, such as genomic instability and bystander effects do not contribute significantly to the interpretation of the epidemiological data.

16 From the report follows that the risks and effects of the dispersal of radioactive material in

the body complex and largely unknown are. UNSCEAR does not use the word ‘radionuclide’ in its text.

17 The studies reviewed by UNSCEAR do not allow a direct estimate of any heritable risks of radiation exposure. Neither do they confirm that there is no risk of heritable effects.

18 UNSCEAR observes that there is a growing evidence of elevated risks of radiation-induced non-cancerous diseases, such as congenital malformations, cardiovascular diseases, cataracts and effects on the immune system.

Apparently the [bevoegdheden] of UNSCEAR to assess all kinds of radiation sources and their effects on the health of populations and individuals are limited. The text of UNSCEAR 2010 Report, written in a bureaucratic language, seems to be [tweeslachtig]. At one hand the Committee [signaleert] studies and new evidence which should be reviewed, at the other hand the Committe seems to stick to 60 years old data bases and seems to be reluctant to endorse scientific efforts to validate new evidence from sources other than than the official United Nations sources.

H.4 WHO

CONSTITUTION
OF THE WORLD HEALTH ORGANIZATION[1]

THE STATES Parties to this Constitution declare, in conformity with the Charter of the United Nations, that the following principles are basic to the happiness, harmonious relations and security of all peoples:

Health is a state of complete physical, mental and social well-being and not merely the absence of disease or infirmity.
The enjoyment of the highest attainable standard of health is one of the fundamental rights of every human being without distinction of race, religion, political belief, economic or social condition.
The health of all peoples is fundamental to the attainment of peace and security and is dependent upon the fullest co-operation of individuals and States.
The achievement of any State in the promotion and protection of health is of value to all.
Unequal development in different countries in the promotion of health and control of disease, especially communicable disease, is a common danger.
Healthy development of the child is of basic importance; the ability to live harmoniously in a changing total environment is essential to such development.
The extension to all peoples of the benefits of medical, psychological and related knowledge is essential to the fullest attainment of health.
Informed opinion and active co-operation on the part of the public are of the utmost importance in the improvement of the health of the people.
Governments have a responsibility for the health of their peoples which can be fulfilled only by the provision of adequate health and social measures.

ACCEPTING THESE PRINCIPLES, and for the purpose of co-operation among themselves and with others to promote and protect the health of all peoples, the Contracting Parties agree to the present Constitution and hereby establish the World Health Organization as a specialized agency within the terms of Article 57 of the Charter of the United Nations.

[1] The Constitution was adopted by the International Health Conference held in New York from 19 June to 22 July 1946, signed on 22 July 1946 by the representatives of 61 States (Off. Rec. Wld Hlth Org., 2, 100), and entered into force on 7 April 1948. Amendments adopted by the Twenty-sixth, Twenty-ninth, Thirty-ninth and Fifty-first World Health Assemblies (resolutions WHA26.37, WHA29.38, WHA39.6 and WHA51.23) came into force on 3 February 1977, 20 January 1984, 11 July 1994 and 15 September 2005 respectively and are incorporated in the present text.

CHAPTER I – OBJECTIVE

Article 1

The objective of the World Health Organization (hereinafter called the Organization) shall be the attainment by all peoples of the highest possible level of health.

p.62

AGREEMENT BETWEEN THE INTERNATIONAL ATOMIC ENERGY AGENCY AND THE WORLD HEALTH ORGANIZATION1

Article I – Co-operation and Consultation

1. The International Atomic Energy Agency and the World Health Organization agree that, with a view to facilitating the effective attainment of the objectives set forth in their respective constitutional instruments, within the general framework established by the Charter of the United Nations, they will act in close co-operation with each other and will consult each other regularly in regard to matters of common interest.

2. In particular, and in accordance with the Constitution of the World Health Organization and the Statute of the International Atomic Energy Agency and its agreement with the United Nations together with the exchange of letters related thereto, and taking into account the respective co-ordinating responsibilities of both organizations, it is recognized by the World Health Organization that the International Atomic Energy Agency has the primary responsibility for encouraging, assisting and co-ordinating research on, and development and practical application of, atomic energy for peaceful uses throughout the world without prejudice to the right of the World Health Organization to concern itself with promoting, developing, assisting, and co-ordinating international health work, including research, in all its aspects.

3. Whenever either organization proposes to initiate a programme or activity on a subject in which the other organization has or may have a substantial interest, the first party shall consult the other with a view to adjusting the matter by mutual agreement.

from: [WHO 2009] Q562