Reprocessing of spent nuclear fuel

Jan Willem Storm van Leeuwen
independent consultant

member of the Nuclear Consulting Group

November 2019
storm@ceedata.nl

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

Contents

TABLES

FIGURES

Roots of reprocessing

Reprocessing technology has been developed during the late 1940s and during the Cold War to recover plutonium from spent fuel from special military reactors for the production of nuclear weapons. In the 1960s and 1970s civil applications of the military reprocessing technology were developed, when de breeder concept came into the picture.

Main purpose of the civil reprocessing plants at La Hague in France and Sellafield in Great Brittain was to recover the plutonium and unused uranium from spent fuel from conventional nuclear power plants with light-water reactors (LWRs), in the belief that uranium and plutonium recycling in the breeder cycle (LMFBR: Liquid-Metal-cooled Fast Breeder Reactor) would soon become the base of civil nuclear power.

The currently most advanced power reactors cannot fission more than 0.5% of the nuclei in natural uranium. The nuclear industry promised (and is still promising) that a closed-cycle reactor system (breeder) could fission 50-100 times more nuclei present in natural uranium, and consequently generate 50-100 times more energy from 1 kg uranium, than the conventional once-through system based on light-water reactors (LWRs). France (*'tout électrique, tout nucléaire'*) and the UK (*'too cheap to meter'*) embarked at the time (early 1970s) on the materialization of the breeder concept, expecting that this could make their energy supply largely independent of fossil fuels within a few decades.

The world has nearly 30 plants for spent fuel reprocessing, according to [ENS 2016] Q667 and [nucl-reproc-wiki 2016] Q668, with nominal processing capacities vaying from 1 Mg used uranium per year to about 3000 Mg U/a. Many of these plants, military installations from the Cold War or experimental plants from the 1960s, are shut down, but none is decommissioned yet. Some 12 reprocessing plants are still operating.

Country	Location	Capacity tU/a	Commissioning or operating period
B	Mol	60	1966-1974
D	Karlsruhe	35	1971-1990
F	Marcoule, UP 1	600	1958-1997
F	La Hague, UP 2	800	1966-1974
F	La Hague, UP 2-400	400	1976-2003
F	La Hague, UP 2-800	1,000	1996
F	La Hague, UP 3	1,000	1990
GB	Windscale	300/750	1951-1964
GB	Sellafield, Magnox	1,500	1964
GB	Dounreay	8	1980-1998
GB	Sellafield, THORP	900	1994
IND	Trombay	60	1965
IND	Tarapur	100	1982
IND	Kalpakkam	100	1998
J	Tokai Mura	90	1977-2006
J	Rokkashomura	800	2006/2007
RUS	Mayak B*	400	1948-196?
RUS	Tscheljabinsk	400	1971
RUS	Krasnojark	800	
USA	Hanford, T-Plant *		1945-1956
USA	Hanford, B-Plant *	1 t/d	1945-1957
USA	Hanford, REDOX *	15 t/d	1952-1967
USA	Hanford, PUREX *	2,400	1956-1972/1983-1988
USA	Savannah, River Site *	~ 3,000	1952-2002
USA	West Valley	300	1966-1972

Table 1

Reprocessing plants, world-wide, * only military use. Source: [ENS 2016] Q667. Not listed here are reprocessing plants in China and Pakistan, see [nucl-reproc-wiki 2016] Q668.

In 1977 President Jimmy Carter banned the reprocessing of commercial reactor spent nuclear fuel in the USA. The key issue driving this policy was the serious threat of nuclear weapons proliferation by diversion of plutonium from the civilian fuel cycle, and to encourage other nations to follow the USA lead. President Reagan lifted the ban in 1981, but did not provide the substantial subsidy that would have been necessary to start up commercial reprocessing. Up until this moment no civil reprocessing occurs in the USA. Military reprocessing plants are still operating in the USA.

Several countries operating nuclear power plants have chosen for the reprocessing option, but is unclear to which extent civil spent fuel is actually reprocessed. In Europe two large reprocessing plants are operating: Sellafield in the United Kingdom and La Hague in France. China, India, Japan, Pakistan and Russia have their own reprocessing plants.

A number of countries has chosen for direct disposal of spent fuel: Canada, Finland, Germany, Spain and Sweden.

When it became evident – though not admitted by the nuclear industry – that the breeder cycle would be technically infeasible, reprocessing became essentially superfluous. The nuclear industry quietly switched to other arguments to justify the exceedingly high investments of a reprocessing plant (counted in tens of billions of euros). Now the *raison d'être* of reprocessing is said to be:

- recovery of the unused uranium from spent fuel for recycling in new nuclear fuel,
- recovery of the plutonium from spent fuel for use in LWR's in MOX fuel and so increasing the retrievable energy content a given mass of uranium,
- volume reduction of the high-level nuclear waste by vitrification,
- closed-cycle reactors, said to be able to generate about 50 times more energy from a given amount of uranium
- transmutation of long-lived hazardous radionuclides into radionuclides with short radioactive half-lifes by partitioning and transmutation (P&T), especially the minor actinides (MAs)
- use of thorium as fertile material in the thorium232/uranium-233 cycle.

These envisioned applications will be briefly discussed in this chapter. However, reprocessing raises also issues concerning nuclear security, health hazards and safety.

Once-through and closed-cycle systems

The present nuclear power plants operate in the once-through mode, using enriched uranium as nuclear fuel. During the fission process in the reactor the content of fission products increases and the content of fissile nuclei in the fuel decreases. After a certain amount of fissions the fission process cannot be sustained anymore, then the spent nuclear fuel is discharged from the reactor and replaced by fresh enriched uranium fuel. Spent nuclear fuel is not suitable for reuse and is stored in cooling pools to let decay the high level of radioactivity and the coupled residual heat generation. In the once-through mode not more than 0.5% can be fissioned of the nuclei in the natural uranium the enriched uranium (nuclear fuel) is made from.

In order to utilise a higher fraction of the uranium nuclei for energy generation, the spent fuel has to be recycled. Recycling of uranium and other fissile nuclei is only possible after removing the fission products and other nuclei that impede the fission process; this separation process is called reprocessing of spent fuel. Closed-cycle nuclear power systems are are addressed in the following sections, along with some other issues inevitably arising from reprocessing.

Reprocessing of spent nuclear fuel

Reprocessing is a complex sequence of chemical and physical separation processes, aimed at separation of spent nuclear fuel into several fractions:

- unused uranium, only a few percent of the uranium nuclei are fissioned in the reactor
- plutonium, generated in the reactor from uranium by neutron absorption,
- fission products,
- minor actinides, mainly neptunium, americium and curium, also formed from uranium by neutron capture,
- zircalloy cladding hulls of the nuclear fuel elements. highly radioactive by neutron capture.

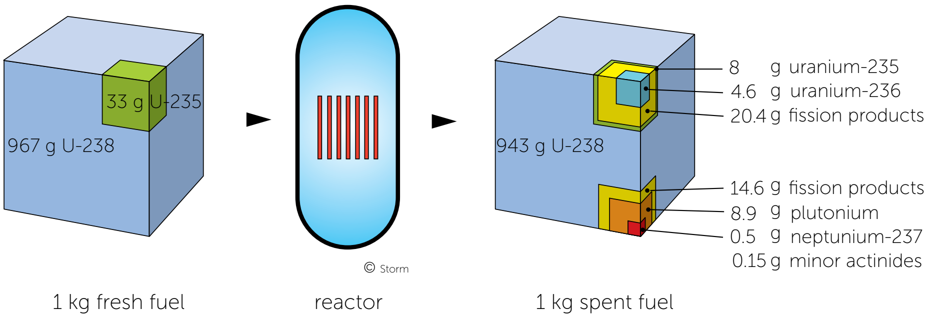

Figure 1

Mass composition of fresh and spent nuclear fuel after three years in a thermal neutron reactor, corresponding with burnup of about 33 GWday/Mg. Source: [Cohen 1977] Q214, [CNRS/EDP 2017] Q744 cites almost the same figures. The amounts of the components other than U-238 in fresh and spent fuel of the newest types of LWRs may be larger, due to a higher burnup of the fuel.

In this diagram the various components of fresh and spent fuel are shown separately. but actually the atoms of the isotopes and elements are dispersed on molecular level. Complete separation is not possible. All new components, represented by the small cubes, are strongly radioactive. The total mass remains nearly constant, a minute fraction is converted into energy during fission. The small cubes at top right represent the mass of the nuclides originating from U-235, the cubes on the lower right corner are formed from U-238.

In addition to the fuel, about 0.6-2 kg zircalloy cladding + spacers and about 25 g silver-indium-cadmium alloy control rods are loaded and discharged with each kilogram of fuel.

In the reprocessing plant the spent fuel elements are chopped into pieces and the contents are dissolved in boiling nitric acid. The resulting solution contains dozens of different elements: fission products, uranium, plutonium and higher actinides. The empty cladding hulls of the chopped and leached fuel pins hardly dissolve and are separated from the solution. Not all fuel dissolves either.

A part of the fission products is gaseous (e.g. noble gases) or easily form gaseous compounds and escape from the liquid. Most of those nuclides are difficult to fix, chemically or physically, into solid materials or containers, notably tritium, carbon-14, iodine-129 and the noble gases such as krypton-85. These nuclides are discharged into the air or sea. In addition a significant fracion of the water-soluble nuclides are discharged with the liquid effluents, notably strontium-90, technetium-99, ruthenium-106 and cesium-137.

Because of the massive releases of radioactive substances into the environment, reprocessing is an exceedingly polluting process. Europe's two operating reprocessing plants, at Sellafield and at La Hague, are situated at the sea coast, for obvious reasons.

Spent fuel is highly radioactive and complicated mixture of many dozens of chemical species: nearly the whole Periodic System of the elements is represented in spent fuel. This results in the occurrence of substantial losses in the separation processes to obtain pure uranium and plutonium and the fact that the recovered metals are unavoidably contaminated with other elements. Process losses and impurities in the products increase with increasing radioactivity of the mixtures.

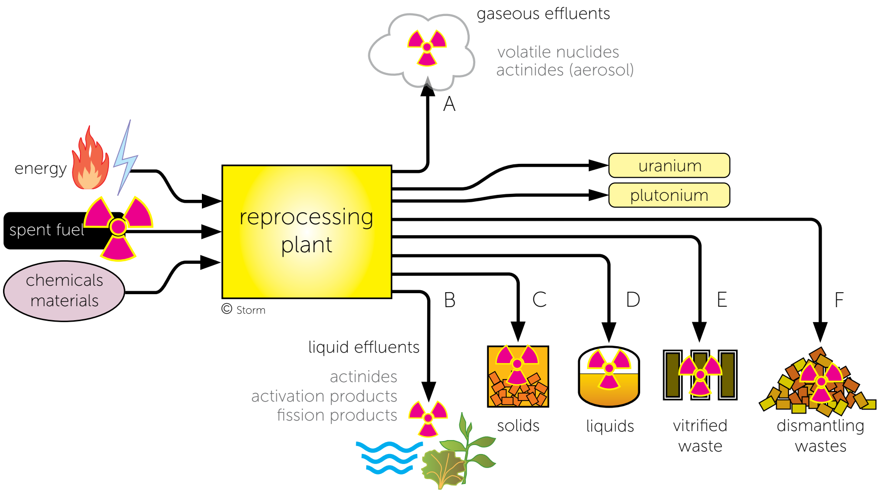

Figure 2

Reprocessing of spent fuel. 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 cannot be vitrified,

C insoluble solid waste consisting of spent fuel cladding hulls, undissolved fuel and and other solids, containing U, Pu, fission products, activation products and actinides, this fraction cannot be vitrified,

D liquid wastes containing fission products, activation products, uranium, plutonium and other actinides, resulting from imperfect separation and purification processes, this fraction cannot be vitrified

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

F An eighth radioactive waste stream, fraction F, consisting of decommissioning and dismantling wastes, will be released after final shutdown of the reprocessing plant, when the plant will be decommissioned and dismantled. This waste consists of decontamination wastes, and radioactive rubble en scrap.

Reprocessing and the Second Law

Separation

Separation processes play a vital role in the process industry, especially in the nuclear process chain. The nuclear process chain starts with the extraction of uranium from its ore, a sequence of physical and chemical separation processes, but also in other processes of the nuclear chain separation processes are important. Separation processes are based on chemical and physical distribution equilibria. These dynamic equilibria are governed by the laws of thermodynamics and never go to completion, as a consequence of the Second Law. For that reason it is impossible to separate a mixture of *n* different chemical species into *n* separate fractions that are 100% pure. There always will be losses and contaminations.

Separation of atomes and molecules implies lowering the entropy of the system: the mixture to be fractionated, and becomes more demanding and goes less completely as:

- more different kinds of species are present in the mixture,
- the concentration of the desirable species in the mixture is/are lower,
- the constituents of the mixture are chemically and/or physically more alike,
- the purity specifications of one or more of the fractions are more stringent,
- the mixture is more radioactive.

Complete separation is a fiction. As a consequence of the above mentioned factors 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 limitations 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.

Purification

Purification of a substance is based on separation processes, aimed at removal of contaminants from the substance. A higher purity means a lower concentration of contaminants. Extracting a species at a lower concentration requires more useful energy and is coupled to greater material losses. Higher purity means better predictable properties of a material. As pointed out above 100% pure materials are impossible. Purity specifications depend on the application of a material. Actually the purity of a material in the process industry is an economic notion.

A single substance can be made more pure at the expense of

- more loss of that substance into the waste streams,
- consumption of more chemicals,
- consumption of larger amounts of energy,
- the need for more complex equipment.

Reprocessed uranium

Recycled uranium, from reprocessed spent fuel, 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.

Above problems worsen each time the fuel passes the fuel cycle. The gamma activity of recycled uranium increases with the number of cycles, because of a growing content of gamma-emitting decay products.

Recycled uranium needs addition of more fissile plutonium than depleted or natural uranium, because of the neutron absorbing properties of U-232, U-234 and U-236. 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 U-232 and U-234 than in U-235 and is enriched also in U-236, 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. In addition the fabrication of fuel elements containing reprocessed uranium is difficult and has to be done by remotely controlled equipment due to its high radioactivity. Utilisation of reprocessed uranium has a negative energy balance, due to high energy consumption of fuel fabrication and the high energy investments of reprocessing of spent fuel including dismantling of the reprocessing plant at the end of its operational lifetime.

The World Nuclear Association [WNA-*mox* 2016] Q246 cites a figure of 45 Gg reprocessed uranium being available for reuse, which would be equivalent to 50 Gg (1 Gg = 1000 Mg) of natural uranium. The displacement figure of 50 Gg is inconsistent with the publication [Foresey & Dickson 1987] Q239. Reprocessed uranium contains slightly more U-235 than natural uranium (around 0.8% vs 0.7%) indeed, but it needs a higher fissile content than equivalent conventional nuclear fuel, as pointed out above.

Evidently this highly radioactive material poses health risks when released into the public domain, by accidents, terroristic actions or otherwise.

Plutonium

Plutonium is generated from uranium-238 (non-fissile) by neutron irradiation in nuclear reactors. The isotopic composition of the plutonium varies with the irradiation time in the reactor. At first the fissile plutonium-239 is formed and from this isotope heavier isotopes are formed by subsequent neutron captures: Pu-240 (non-fissile), Pu-241 (fissile) and Pu-242 (non-fissile). In nuclear fuel at low burnup little Pu-239 is transformed into heavier isotopes. The higher burnup of the fuel, the longer the stay time in de reactor and the more non-fissile heavy plutonium isotopes are generated.

Weapons-grade plutonium contains typically 93.6% Pu-239 [O'Connor 2003] Q599 is produced by neutron irradiation of uranium in special military reactors. The nuclear fuel from these reactors has a very low burnup (about 100 MW(th).days/Mg) before reprocessing, so only small amounts of the higher isotopes of plutonium (e.g. the non-fissile isotope Pu-240) are formed. Higher isotopes of plutonium and trans-plutonium elements make plutonium more radioactive and less suitable for production of nuclear weapons.

Reactor-grade plutonium originates from spent fuel from civil power reactors and contains typically less than 65% fissile plutonium isotopes (Pu-239 + Pu-241). In commercial reactors the fuel elements stay far longer and get a higher burnup (33000-46000 MW(th).days/Mg) than in military reactors. Due to longer stay times in the reactor, more of the heavier plutonium isotopes are formed: Pu-240, Pu-241 and Pu-242, but also Pu-238. The even isotopes are not fissile in LWR's and in bombs. Moreover, trans-plutonium elements, e.g. Am-241, Am-243, Cm-244, are formed from plutonium isotopes by neutron capture. According to [WNA-*pu* 2016] Q247:

> The term 'fissionable' applies to isotopes that can be made to undergo fission. If a fissionable isotope only requires neutrons with low kinetic energy to undergo fission, then it is said to 'fissile'. Thus, all fissile isotopes are fissionable. Pu-240 is fissionable, as it undergoes fission in a fast neutron reactor - but it is not a fissile isotope.

Contrary to statements of the nuclear industry [WNA 2012b] Q541 reactor-grade plutonium is suitable for nuclear explosives, according to [Barnaby 2005a] Q339 and [Barnaby 2005b] Q240, [Glaser 2005] Q593, [Schneider 2007] Q590.
Plutonium has a much lower critical mass than uranium. The bare-sphere critical mass of weapons-grade plutonium is 11.5 kg (diameter 10.5 cm) and of reactor-grade plutonium 14.6 kg (diameter 11.5 cm). With a neutron reflector of 15 cm the figures are: 3.71 kg (7.20 cm), respectively 4.58 kg (7.72 cm), according to [Glaser 2005] Q593.

Pu-238 is a strong alpha emitter. By beta decay, plutonium-241 is transformed into americium-241; Am-241 is a strong gamma emitter, greatly increasing the gamma activity of the plutonium. Within a few years storage time, the concentration of Am-241 builds up to a level the plutonium cannot be handled safely anymore. With a content of Am-241 higher than 1% it has to be purified again [Hulst & Mostert 1979] Q242, a costly process. For recycled plutonium from LWR with MOX fuel, the repurifying limit due to Am-241 may be reached about one year after reprocessing. Americium-241 decays to neptunium-237, a fissile nuclide.
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. The same holds true for the burnup of the fuel from which the plutonium is extracted: the higher the burnup, the longer its stay in the reactor and consequently the less the isotopic quality of the plutonium.

Neptunium-237

During the fission process in the reactor the short-lived neptunium-239 isotope is formed from uranium-238 by neutron capture. Neptunium-239 decays with a half-life of 2.35 days to plutonium-239, a first-rate bomb material. In addition sizeable quantities of the long-lived isotope neptunium-237 (half-life 2.14 million years) are formed, mainly by neutron capture of uranium-235 and decay of americium-241. According to [KfK 1983] Q587 roughly 400-700 g Np-237 per metric ton spent fuel are formed, depending on the burnup of the nuclear fuel. Np-237 can be separated by chemically means from the other elements in spent fuel, due to its specific chemical properties: it is a separate chemical element. After a cooling period of less than a year Np-237 is the only neptunium isotope remaining in spent fuel. Consequently it is possible to obtain a pure fissile material from spent fuel just by chemical means, without enrichment.

Neptunium-237 is fissile and can be used to produce a nuclear explosive device. Its critical mass is comparable to that of uranium-235. One or more nuclear weapon states may have tested a nuclear explosive using Np-237. Historically, neptunium 237 has been separated by the nuclear weapon states in only small quantities, principally for non-explosive uses, as target material for plutonium-238 production. Pu-238 can be used as neutron initiator of nuclear weapons.

By the end of 1997, the world inventory of neptunium and americium was estimated to exceed 80 metric tonnes, or enough for more than 2,000 nuclear weapons, and the amount is growing at a rate of as many as 10 tonnes per year. If actinide separation becomes routine, inventories of separated neptunium-237 and americium will escalate, according to [ISIS 1999] Q552.

Americium

According to [KfK 1983] Q587 about 120 g americium isotopes per metric ton spent fuel are formed at a burnup 33 GWe.day/Mg; at higher burnups the yield is proportionally larger. Americium has to be separated from plutonium and uranium after reprocessing, for reason of the high radioactivity of the americium isotopes and their unfavorable nuclear properties as reactor fuel. Assuming the isotope Am-242 (half-life 16 hours) has already decayed to Cm-242, the main isotopes of americium in spent fuel are Am-241, Am-242m and Am-243. Just like neptunium, americium can be separated by chemically means from the other elements in spent fuel, due to its specific chemical properties.

All americium isotopes are fissile and can be used to produce a nuclear explosive device, so it is possible to obtain undiluted bomb material from spent fuel just by chemical means. Estimates of the bare-sphere critical mass of the americium isotopes vary from 9-150 kg. However, under special conditions the critical mass of Am-242m may be as low as 7 grams, according to [Ronen *et al*. 2000] Q243.
Historically, americium has been separated by the nuclear weapon states in only small quantities, principally for non-explosive uses: for smoke detectors, neutron generators, and research activities. During reprocessing of spent fuel americium is usually discarded in the high-level waste streams.
The world inventory of Am-241 at the end of 1997 is estimated at some 45 tonnes and is growing by about 4 tonnes/year. This amount of Am-241 is the result of the decay of plutonium-241. In nuclear weapon programs and civil plutonium recycle programs, americium-241 is separated from aging plutonium to purify it and reduce the material handling problems caused by americium’s radioactive emissions [ISIS 1999] Q552. ISIS estimates the worldwide separation of americium at some 100 kg/yr.

MOX fuel in LWRs

The plutonium recovered from spent fuel, usually labeled reactor-grade plutonium Pu_{rg} could be used to fabricate MOX (Mixed OXide) fuel elements consisting of natural or depleted uranium oxide and plutonium oxide instead of a higher content of uranium-235, to be used in light-water reactors (LWRs). At present about 30 nuclear power plants, mostly in Europe, are using MOX fuel elements, not more than about 30% of the core. Special reactor designs are required to replace all enriched uranium fuel in the core fully by MOX fuel. Reprocessing of commercial nuclear fuel to make MOX is done in the United Kingdom and France, and to a lesser extent in Russia, India and Japan [wiki-*mox* 2017] Q741.

Plutonium from reprocessed fuel is usually fabricated into MOX as soon as possible to avoid problems with the decay of short-lived isotopes, in particular Pu-241 that decays (half-life 14.1 years) to americium-241, a strong gamma emitter. Within 5 years typical reactor-grade plutonium would contain too much Am-241 to safely handle [WNA-*mox* 2016] Q246.

How much natural uranium could be displaced by using MOX fuel instead of enriched uranium in nuclear power stations?

According to [WNA-*mox* 2016] the current commercial MOX fuel, equivalent with enriched uranium at 4.2% U-235, has an average plutonium content of 9.5%, containing 65% fissile plutonium (Pu-239 + Pu-241). This study assumes that depleted uranium is used for the MOX fuel. The fissile content of U-235 (about 0.3%) of the depleted uranium is ignored for convenience.

Based on these figures 1 Mg reactor-grade plutonium would correspond with 10.5 Mg MOX, of which 9.5 Mg depleted uranium. To produce an equivalent amount of enriched uranium (4.2% U-235) about 100 Mg natural uranium would be needed, assumed a feed/product ratio of 9.5 and 0.3% U-235 tails assay of the enrichment. So 1 Mg Pu_{rg} could save 100 Mg U_{nat}.

[WNA-*mox* 2016] states that the present global inventory of reactor-grade plutonium available for commercial reactors is 320 Mg and that this amount could save 60000 Mg natural uranium. According to the estimate of this study 320 Mg Pu_{rg} would be equivalent to 32000 U_{nat}. This is about one half of the current world annual consumption of U_{nat}. It is unclear how the World Nuclear Asdsociation derived its estimate.

If all spent fuel of the global nuclear fleet would be reprocessed some 60 Mg/year of Pu_{rg} would become available for MOX fabrication. This could save about 6000 Mg/ year of U_{nat} less than 10% of the current annual consumption.

During burning of MOX the ratio of fissile (odd numbered) isotopes to non-fissile (even) drops from around 65% to 20% depending on burnup [wiki-*mox* 2017] Q741. For that reason used MOX fuel is not reprocessed.

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. Serious criticallity problems highly 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. 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 (see above).

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.

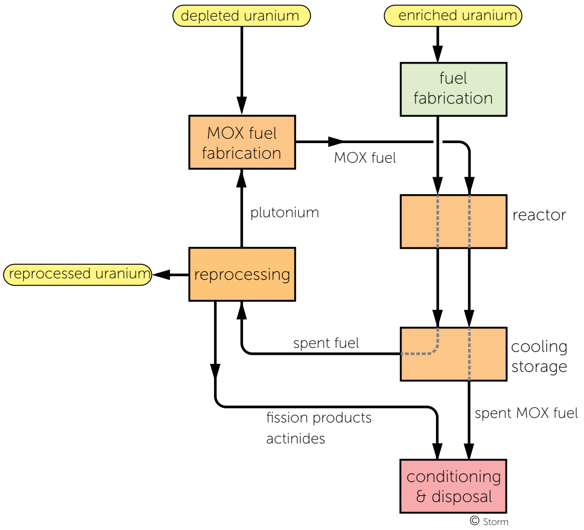

Figure 3
Outline of plutonium recycling in LWRs. The reactor starts up loaded with conventional enriched uranium fuel. The spent fuel is reprocessed, after a cooling period. The recovered plutonium is used to enrich depleted uranium. The resulting MOX fuel is placed into the reactor. Spent MOX fuel is not reprocessed. The reprocessed uranium is seldom reused, because it has a troublesome isotopic composition and is contaminated by fission products.

The MOX option has a negative energy balance, as other nuclear concepts that depend on reprocessing of spent fuel. It takes more energy to reprocess spent fuel, including a proportional part of the energy requirements of the dismantling of the reprocessing plant, and to fabricate MOX fuel than can be generated from that fuel. Moreover, the use of MOX fuel poses serious proliferation and terroristic hazards, as will be discussed in one of the following sections.

Destruction of HEU and Pu: burning the Cold War legacy

Reactor-grade fuel can be made by blending weapons-grade Pu and Highly Enriched Uranium (HEU) from nuclear weapons, with depleted or natural uranium.
A rough estimate of the contribution to the world energy supply by 'burning' these materials can be made, starting with the following figures:

- depleted uranium: about 1.2 million Mg world wide [WNA inf14 2002] Q244
- weapons-grade plutonium (90% Pu-239), about 260 Mg [WNA inf13 2003] Q245
- Highly Enriched Uranium (HEU), weapons-grade 93% U-235 'diluted' to about 2000 Mg 20% U-235, still called HEU ([WNA inf13 2003] Q245.

For a reference reactor of 1 GW(e) which uses a reload of 20.30 Mg fuel for each 0.82 full-power year, the weapons-grade materials would be sufficient to produce about 725 reloads, assuming the use of depleted uranium, enriched to 5% Pu or 4.2% U-235. This amount of reloads corresponds with about 600 full-power years and would meet the world demand (360 GW(e) in 2002) for about 1.7 years. To produce this fuel, about 14000 Mg depleted uranium would be needed.

In addition to the military plutonium, reactor-grade plutonium (typically less than 70% fissile) from the civil reprocessing plants is available. An amount of 400 Mg would be sufficient for about 280 MOX reloads with an enrichment of 7% Pu [WNA inf29 2003] Q246 and [WNA inf15 2002] Q247. This would meet the world demand for about 0.6 years. A mass of about 5300 Mg depleted uranium would be used in this MOX fuel. Together, the military materials and the civil plutonium would be sufficient to meet the present needs of the world nuclear fuel consumption for about 2.3 years, using nearly 20000 Mg depleted uranium.

Figuring the energy consumption and production of the processes involved, fuelling reactors with Pu and HEU would not add energy resources in the simply way as it may look, because the energy consumption in obtaining the Pu and HEU has to be taken into account.
Use of depleted uranium would add some energy resources because the energy consumption of its production is taken into account in the HEU and Pu production. The net energy result of the use of Pu, HEU and depleted uranium probably is not significant, or may be even negative. Moreover, on global scale the contribution would be marginal, even when the energy consumption of the Pu and HEU production is neglected.

Uranium-plutonium breeder systems

In its publication *Radioactive Wastes – Myths and Realities* [WNA 2016c] Q542 the World Nuclear Association states:

> The fuel for nuclear power is virtually unlimited, considering both geological and technological aspects. There is plenty of uranium in the Earth's crust and furthermore, well-proven (but not yet fully economic) technology means that we can extract about 60 times as much energy from it as we do today.

Obviously this statement refers to the closed-cycle technology, the breeder system, that would generate more fissile nuclides from fertile nuclides than it consumes.

What is called a 'fast reactor' or breeder is not just a reactor but a cyclic system consisting of a fast-neutron nuclear reactor plus reprocessing plant plus fuel fabrication plant. Each of the three components of the breeding cycle would have to operate flawlessly and finely tuned to each other for decades without interruption. If one component fails in any respect, the whole system fails and breeding is out of question. Operation of the cyclic system is further complicated by the high radioactivity of the materials to be processed, increasing with each following cycle.

Decades of intensive research in several countries (e.g. USA, UK, France, Germany, the former Soviet Union) and investments of some $100bn, have proved that the breeding cycle is technically infeasible.

The causes of this failure have nothing to do with arguments like: 'not economically attractive' (obviously a technically unfeasible system is not economically attractive) nor with protests of environmental activists. The failure of materialization of the breeder concept can be traced back to fundamental laws of nature, particularly the Second Law of thermodynamics. From this law it follows, among other consequences, that separation processes of mixtures of different substances never go to completion and consequently perfect materials are not possible. From the Second Law it also follows that the deterioration of materials by ageing processes are inevitable.

Pivotal in the breeder cycle is the reprocessing of the spent fuel as soon as possible after unloading from the reactor, to avoid the decay of some isotopes, for example plutonium-241 into americium-241. Due to the rapidly increasing radioactivity of the spent fuel with each cycle, reprocessing and fuel fabrication become increasingly difficult. The isotopic compositions of the recovered uranium and plutonium become less favourable each cycle. Due to the unavoidable and increasing separation losses, the cycle produces less fissile nuclides than it consumes. For these reasons, among other, the breeder cycle is technically infeasible.

Summarised, the breeder concept is inherently unfeasible, because is implicitely based on infeasible assumptions:

- availability of perfect materials, not subject to spontaneous degrading processes (ageing)
- technical systems with 100% predictable properties and behavior across decades
- 100% perfect separation of a mixture of a large number of different chemical species into pure fractions, regardless of the radioactivity of the mixture.

All three conditions are inconflict with the Second Law of thermodynamics.

This conclusion is also valid for the proposed thorium breeder cycle and the partitioning & transmutation (P&T) system, both to be discussed in the following sections.

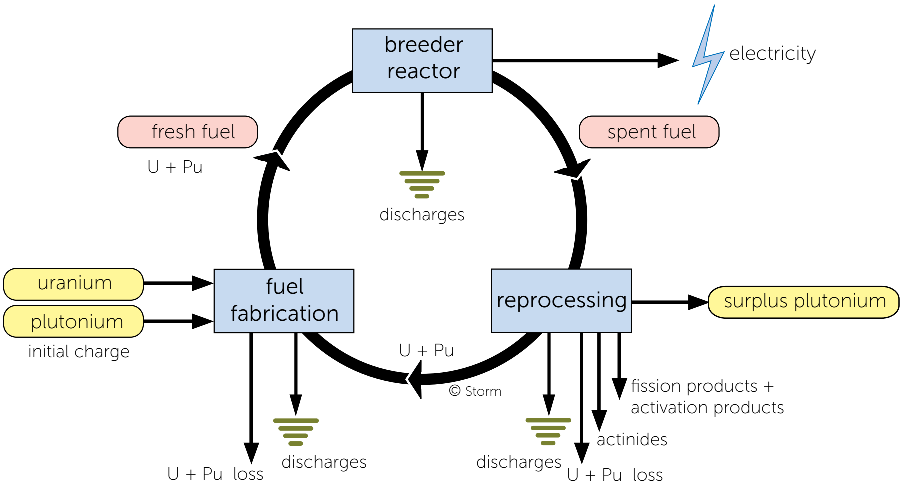

Figure 4
Outline of the concept of the breeder system in steady state. By repeatedly recycling spent fuel, it would be theoretically possible to fission the main part of natural uranium. If all goes well, the cycle produces during its operational life a plutonium gain, large enough to start up two or more new breeders: one to replace the closed down unit, and one or more additional breeders. The cycle represents the mass flows of uranium and the nuclides originating from the nuclear processes in the reactor (fission, activation and decay). The initial plutonium charge to start up the breeder reactor is about 3 Mg Pu for a 1 GW(e) FBR.

Reprocessing of FBR fuel

Reprocessing of Fast Breeder Reactor (FBR) 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 FBR fuel is much stronger radioactive than spent LWR fuel.

High burnup and the high Pu content (15-20 times greater than that of irradiated LWR fuel) 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.
- More unsoluble compounds and noble metal alloys (Ru, Rh, Tc, Mo, Pd), possibly containing plutonium, and undissolved MOX particles remain in the dissolver. The undissovedparticles 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 actindes 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 the FBR fuel reprocessing plant would benefit less from advantage of scale than a LWR fuel reprocessing plant.

Some other difficulties, specific for spent FBR fuel are:
The fuel assemblies of a FBR, made of stainless steel, have to be disassembled before the fuel pins can be chopped into smalle pieces. Because of the high radioactivity and heat output of the irradiated fuel and 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.

Discharges to the environment (water and air) of actinides may be higher than in a LWR cycle, because of the higher actinide content in FBR fuel.
Costs of FBR fuel reprocessing could be at least twice as much as that of LWR fuel [UNIPEDE/CEC 1981] Q58 .

Thorium for fission power

Thorium is a radioactive metal, more abundant in the Earth's crust than uranium. The concept of the thorium reactor is based on the conversion by neutron capture of non-fissile thorium-232 into uranium-233, which is as fissile as plutonium-239. In common with the uranium-plutonium breeder the thorium-uranium breeder is not just an advanced reactor, it is an intricate cyclic system of reactor, reprocessing plant and fuel element fabrication plant. Each of the three components of the cycle has to operate flawlessly for decades, finely tuned to the two other components.

The feasibility of the thorium breeder system is even more remote than that of the U-Pu breeder. After four decades of research there are still no solutions for the basic problems mentioned by [ORNL-*5388* 1978] Q376. The fundamental obstacles that render the U-Pu breeder technically unfeasible apply also to the thorium breeder. A major drawback of the thorium cycle is that a thorium reactor cannot sustain a fission process in combination with breeding uranium-233 from thorium-232, but will always need an external accelerator-driven neutron source, or the addition of extra fissile material, such as plutonium or uranium-235 from conventional reactors.

Building up a Th-U-233 breeder system would pose a severe logistic problem, even if the system would work as advertised. Only small quantities of U-233 exist in the world at this moment, the USA has 1710 kg of it in storage, 905 kg of which still contained in spent fuel. The U-233 stocks in other countries are unknown. The largest DOE reactor currently operating could produce only about 0.3 kg/year.

It would take decades to obtain sufficient U-233 from special reactors to start up the first operating Th-232–U-233 breeder system. After that it would take more than 8 doubling times to attain a thorium breeder capacity equalling the current nuclear capacity (about 370 GW). Even with an assumed unrealistically short doubling time of 20 years more than 8 doubling times would mean nearly two centuries.

Among a number of other countries, the USA conducted Th-232/U-233 research in the 1960s and 1970s (e.g. in the Shippingport reactor), the research has not been continued. U-233 has been envisioned as fuel in very compact military reactors for special applications, and in spacecraft. Although U-233 reportedly would have about the same properties as plutonium for use in nuclear weapons, no such weapons seem to have been developed. Apparently there exist good reasons not to use U-233 in military reactors and weapons and not to continue the research towards the thorium power reactor. India seems to be the only country at this moment still conducting some research on Th-232/U-233 fuel cycle. An overview of research projects in the past and of advanced thorium reactor concepts is given in [WNA-*Th* 2015] Q302.

The realisation of the thorium-U-233 breeder cycle has a number of hurdles to overcome according to [ORNL-*5388* 1978] Q376, [ORNL-*6952* 1999] Q377 and [PSR-IEER 2009] Q617 such as:

- the separation processes needed to recycle fissionable material are inherently incomplete, so significant losses are unavoidable
- the recycling of Th-232/U-233 fuel has yet to be demonstrated
- assumed the recycling would be technically feasible, it is still unknown if the cycle would produce sufficient U-233 to expand the Th-232/U-233 capacity, or even to maintain itself
- the radioactivity of the spent fuel and recycled U-233 increases with every cycle, while its isotopic quality decreases, consequently its usefulness as fissile material decreases
- due to the increasing radioactivity the separation processes deteriorate and the separation will get even more incomplete
- due to the increasing radioactivity the fuel handling and fresh fuel fabrication becomes increasingly difficult.

Since the publication of the above mentioned ORNL reports no publications are found reporting a solution to each of these challenges, a *conditio sine qua non* for materialisation of the thorium-U-233 breeder cycle.

Uranium-233

Uranium-233 is a fissile nuclide that is prepared from non-fissile thorium -232 by neutron irradiation in a nuclear reactor. After irradiation the thorium target elements are to be reprocessed to separate the U-233 from the remaining Th-232. U-233 has been used during the 1950s and 1960s in the development of nuclear rockets, nuclear ramjets for an atomic bomber, but also for civil power reactors. These technical developments were halted in the 1970s, apparently due to various problems. One of these problems is the presence of uranium-232, a strong gamma-emitter, which makes U-233 difficult to handle. Methods to limit the content of U-232 are expensive.
Uranium-233 has a critical mass much less than U-235 and is comparable to plutonium in terms of weapons-usability. Between 1955 and 1968 several nuclear weapons test were conducted using uranium-233 [Alvarez 2012] Q594.
In the United States about 1550 kg of U-233 was separated. Of this amount about 123 kg may be unaccounted for, enough for some 13 nuclear explosive devices. The radiation level from contaminants is not considered to be an adequate barrier to prevent a terrorist from making an improvised nuclear device. Storage of the US stockpile of U-233 is a safeguard, security and safety risk. The production of the stockpile also has left a disposal burden [Alvarez 2012] Q594.

How is the situation concerning U-233 in elsewhere in the world? Some countries may be still involved in the development of a thorium-232/uranium-233 nuclear breeder system.

Molten salt reactor

In many publications on thorium-fuelled reactors and on partioning & transmutation systems the concept of the Molten Salt Reactor (MSR) comes up. In this type reactor the nuclear fuel is dissolved in a mixture of molten fluoride salts at high temperatures. Advocated advantages of the MSR would be: compactness, inherent safety, operation at atmospheric pressure and high operating temperatures in the range of 600-900 °C. Fast neutron and thermal neutron designs are proposed, in once-through as well in breeder modes.

The concept of the MSR was established in the 1950s. As far as known only three MSRs are ever built, and that went critical, according to [wiki-*msr* 2017] Q693, all three in the USA:

- Aircraft Reactor Experiment (ARE), designed for use as an engine in a nuclear-powered bomber. Power 2.5 MWth, peak temperature 860 °C, produced 100 MWh in nine days in 1954.
- Pratt & Whitney Aircraft Reactor-1 (PWAR-1); the experiment run for a few weeks in 1957, the reactor was critical at zero nuclear power, temperature 675 °C.
- Molten-Salt Reactor Experiment (MSRE), operated at a power of 7.4 MWth and temperature of 650 °C, during the period 1965-1969.

As of 2011 the ARE and MSRE remained the only MSRs ever operated. After these experiments a number of research projects in various countries were set up, among other thorium breeder designs, see the overviews in [wiki-*msr* 2017] Q693 and [WNA-*msr* 2016] Q739. As far as known none of those designs after 1969 ever were built and operated.

From the above observations may be concluded that the promising MSR concept is not viable. Doubtless Second Law phenomena are playing an important part in the MSR demise.

Partitioning & transmutation

Partitioning & transmutation (P&T) is a technologic concept proposed by the nuclear industry as a means of 'destroying' long-lived radionuclides. If the concentrations of the long-lived dangerous radionuclides in the nuclear wastes could be reduced to below an official standard (which standard is not indicated), the waste would be safe for release into the public domain after a storage time of 'only a few' centuries to a thousand years, instead of 100000s 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.
Some publicaties suggest even the possibility of nuclear energy with (almost) no radioactive waste for example [LPSC 2001] Q451 and [ORNL 2013] Q539.

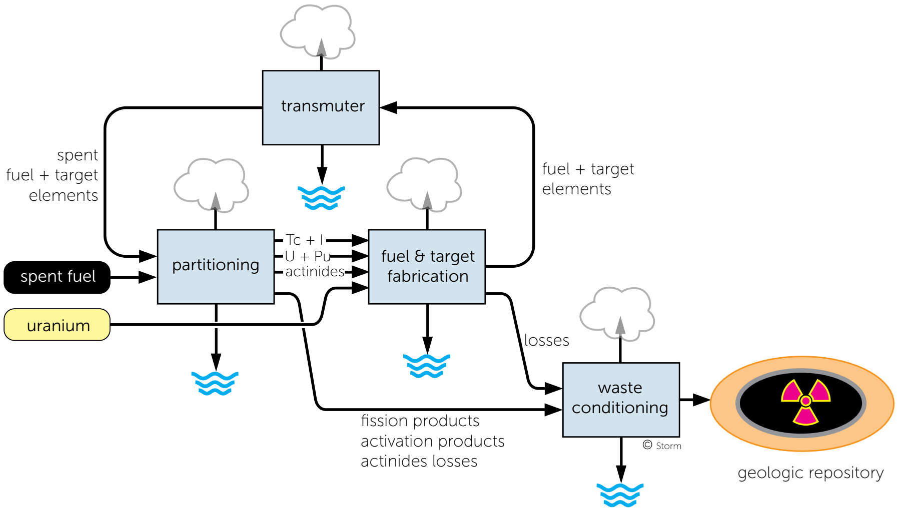

Figure 5
Principle of a partitioning & transmutation system. All material flows indicated by the arrows and all discharges into the biosphere are hightly radioactive,

A partitioning & transmutation system is a cyclic system consisting of three components: a transmuter reactor, a partioning plant and fuel fabrication plant. Each of the three components of the cycle would have to operate flawlessly and finely tuned to each other for decades without interruption, if one component fails in any respect, the whole system fails.
In the partitioning plant, a more advanced reprocessing plant, spent fuel would be separated into more partitions than in a conventional reprocessing plant. For this reason partioning is more demanding and more energy-intensive than conventional reprocessing.
Some partitions, especially the minor actinides (MAs), would be incorporated in special fuel and target elements. Due to the high level of radioactivity fuel and target fabrication is a demanding process, to be done by robotic equipment.
In the transmuter reactor the long-lived adionuclides would be transmuted into shorter-lived radionuclides by neutron capture. The neutrons are to be produced by fission of fissile nuclei in the mixture, and/or by an external source.

Three principal transmuter reactor 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. Other transmuter concepts are the PBR (particle bed reactor or pebble bed reactor), a helium-cooled fast reactor, and molten salt reactors (MSRs), see for example [WNA-*reproc* 2016] Q738 and [WNA-*ane* 2016] Q740.

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 P&T 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.

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-*2897* 1970] Q254, [Fischer 1986] Q240 and [Roepenack *et al.* 1987] Q241. Evidently these long-known facts cause mounting difficulties in the handling of the recycled uranium and plutonium. 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.
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.

According to the Swedish study [SKB 2010] Q447 the development of a functioning P&T 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, assumed these systems woulld be feasible. To fission the existing amounts of actinides would require the perfect functioning of a number of P&T systems during at least one century and probably longer [SKB 2010].

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.

Other conclusions are formulated by [Pistner *et al*. 2015] Q734, such as:
- Partitioning and transmutation of actinides does not result in a relevant reduction of dose rates from final repositories, despite the huge efforts and costs involved, as the dose rates are dominated by nuclides such as I-129, Se-79, Cl-36, C-14 that will not be separated according to current P&T concepts.
- P&T does not reduce timelines for safe isolation of the waste, as dose rates are dominated with long-lived mobile fission and activation products, that are not influence by a P&T treatment.
- Recycling is not possible with some exceptions.
- Interim storage is not sustainable in the long term.
- Transmute would have high costs and sophistication, long duration, low benefit with respect to final disposal of high level wastes.
- Deep geologic repository is the best currently available option.

Apart from the demanding properties of the transmuter reactor, partitioning and fuel/target fabrication are two very energy-intensive processes. 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 would require very high energy investments, due to the highly radioactive construction materials. The P&T system would be an energy sink to such extent that a nuclear energy system consisting of a number of nuclear power plants coupled to associated P&T systems might be an energy sink instead of a net energy source.

The P&T cycle (Figure 5) would be an exceedingly polluting system. The material flows in the cycle are strongly radioactive, even more than in the breeder cycle, and discharges into the human environment by each component of the cycle would be unavoidable, caused by Second law phenomena. Due to the fact that relatively large amounts of minor actinides, that are highly radiotoxic, would circulate in the system, discharges and leaks would pose serious health hazards. Even without accidents the pollution by long-lived radioactive materials would become considerable.

Accidents could have very serious consequences and the chance of severe accidents might be more serious than in the conventional nuclear power plants due to the presence of large amounts of strongly radioactive materials in mobile form at different locations during decades or even centuries.

Another point of concern would be increased risks of nuclear terrorism for the same reasons. Substantial amounts of fissile materials would be circulating in the P&T cycle. The risks of dirty bombs could also increase with time as a consequence of the increasing amounts of radioactive materials in mobile form with time.

Partitioning of spent fuel, separation into a number of fractions , is prerequisite for a P&T system would be even more demanding than reprocessing in the breeder cycle. Materialisation of a P&T system is infeasible for it is based on assumptions that are in conflict with the Second Law, the same as discussed in the sections on the breeder cycle and thorium reactor.

Nuclear terrorism

MOX is the acronym of Mixed OXide fuel, nuclear fuel with plutonium instead of U-235. MOX fuel is relatively little radioactive and can be handled without specialized equipment. A terrorist group would have little difficulty in making a crude atomic bomb from MOX fuel. Separating uranium dioxide and plutonium dioxide from MOX fuel can be done using straightforward chemistry. Converting the plutonium dioxide into plutonium metal, and assembling the metal together with conventional explosives to produce a crude nuclear explosive does not require materials from special suppliers. The information required to carry out these operations is available in the open literature [Barnaby 2005a] Q339, [Barnaby 2005b] Q340. Technology needed to make nuclear bombs from fissile material is available outside of the established nuclear-armed countries and in the open literature, as proven in '*Nth Country Experiment*' [Frank 1967] Q591, [Schneider 2007] Q590.
The authors of [MIT 2003-2009] Q280 considered the proliferation and safety risks of reprocessing and the use of mixed-oxide (MOX) fuel unjustified. But there are also economic reasons not to recycle in their view.
Studies by the Oxford Research Group show that MOX fuel poses a large and underrated terrorist risk [Barnaby & Kemp 2007] Q360. The 6 kg of plutonium contained in the Nagasaki bomb would fit in a soft drink can.

Nuclear weapons can be made from reactor-grade plutonium, as pointed out above, although those made using weapons-grade plutonium are more effective. The USA and UK exploded devices based on reactor-grade plutonium in 1956 and in the 1960s. A good nuclear weapons designer could construct a nuclear weapon from 4-5 kg of reactor-grade plutonium. Less reliability or a less predictable explosive yield than a military weapon would not be a problem for a terrorist group planning an attack in the center of a large town. This is the reason why so many scientists all over the world are strongly opposing the reprocessing of spent fuel and the use of MOX fuel in civilian reactors.

A considerable part of nuclear security problems concerning fissile materials suitable to make crude nuclear explosives, plutonium, neptunium and americium, originate from one source: reprocessing of civil spent fuel. In addition uranium-233 is recovered by reprocessing spent fuel from special thorium-uranium reactors.
Do the benefits of reprocessing outweigh the security and health risks it generates plus the costs of safeguarding the separated dangerous materials?
Without reprocessing the only way to acquire fissile bomb material would be enrichment of uranium.

Vitrification

According to [WNA 2012b] Q541 a typical 1 GWe reactor produces each year about 700 kg high-level wastes, contained in about 23 Mg (metric tons) of spent fuel. With 'high-level wastes' WNA likely refers to the fission products plus actinides in spent fuel, but does not mention them explicitely. After separation from spent fuel in a reprocessing plant the liquid high-level wastes are evaporated to solids, mixed with glass-forming materials, melted and poured into stainless steel canisters which are then sealed by welding. The vitrified waste from the operation of a 1 GWe reactor for one year would fill about twelve canisters, each 1.3 m high and 0.4 m diameter and holding 400 kg of glass, according to WNA.

The canisters are to be placed in a geological repository for permanent disposal. In another process, called Synroc, the wastes are calcined and mixed wich several metaloxides for conversion at high temperatures into a crystalline ceramic material (synthetic rock).

Applying the vitrification concept the mass holding the highly radioactive materials would be reduced from 23 Mg spent fuel to 4.8 Mg borosilicate glass, a reduction of a factor of less than five.

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

This concept sounds simple and may seem plausible at a first glance. However, on closer examination the feasibility as radioactive waste reduction strategy proves to be based on fallacies and ignorance of the Second Law of thermodynamics.

Long-lived fission products and the minor actinides are considered to be the most dangerous radionuclides in spent fuel. The idea behind the vitrification concept is to separate these dangerous radionuclides from spent fuel and to chemically immobilise them in a matrix of borosilicate glass. The fission products and actinides (other than U and Pu) then are converted into oxides, which are mixed with a glassmaking frit and melted to form a borosiicate glass. The molten glass is poured into appropiate stainless steel containers, which are to be placed in a geological repository for permanent disposal. The remaining radioactive wastes are considered to be not dangerous, because of a lower specific activity, and could be disposed of in a less expensive way, a routine job according to MacKay.

As a means of volume reduction of high-level waste the vitrification concept turns out to be a fallacy: the radioactive waste volumes increase enormously by reprocessing, as will explained below.

By reprocessing the radioactive contents of the spent fuel are distributed among large volumes of different materials, and only a part of it ends up in the vitrified waste, see Figure 6. This greatly enhances the chances of dispersal and of severe accidents involving massive amounts of radioactivity. A significant part of the radioactive contents is released into the environment.

In its communication to the public on the waste vitrification concept the nuclear industry does not pay attention to some practical aspects of the proposed technique, such as:

- All gaseous fission products and a substantial fraction of other fission products and of the actinides are inevitably discharged into the environment by the reprocessing plant.
- Generation of large volumes of other radioactive wastes during reprocessing because substantial fractions of the radionuclides from spent fuel are spread among non-radioactive solids and liquids and do not end up in the glass. Actually the radioactive waste volume increases to a great multiple of the volume of spent fuel.
- Immense amounts of radioactive waste, contaminated by fission products and actinides, will result from decommissioning and dismantling of nuclear power plants and reprocessing plants.
- Some nuclides are discharged during solidifying the liquid waste stream (calcination) and subsequent vitrification of the solid residu. Not all nuclides can be effectively fixed into a glass, such as: Se-79, Ru-106, I-129, since they hardly form stable compounds with the borosilicate matrix, or become volatile during the calcination process.

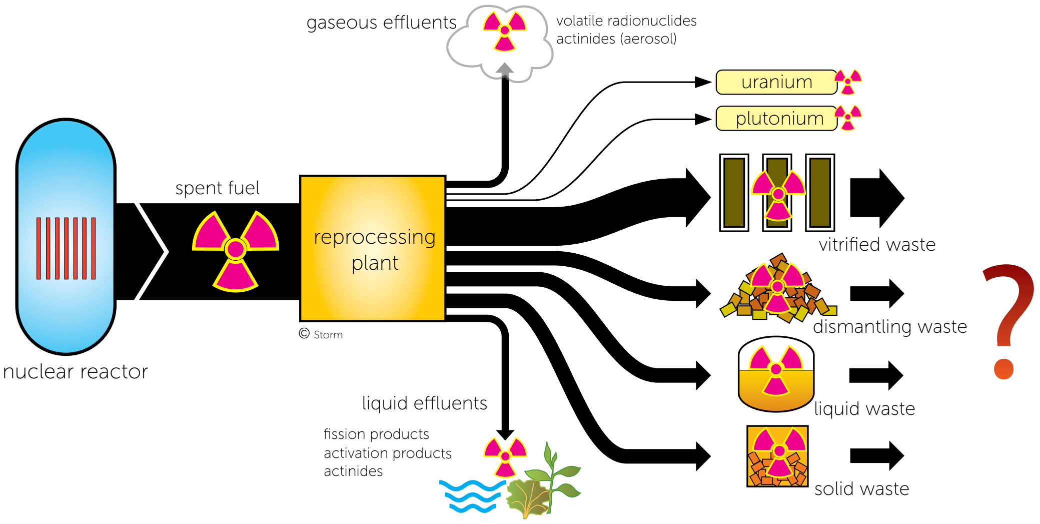

Figure 6

Redistribution of the radioactive contents of spent fuel in the waste streams from reprocessing. In order to recover uranium and plutonium from spent fuel, the radioactivity from the spent fuel is partitioned into a number waste streams. Significant amounts of radionuclides are discharged into the environment. Only a part of the radionuclides from the spent fuel can be vitrified.
One radioactive waste stream, consisting of dismantling wastes, will be released after final shutdown of the reprocessing plant, when the plant is decommissioned, cleaned up and dismantled.
How to safely isolate the radioactive waste streams from the biosphere is still an open question.

The assumption that 100% complete separation of all chemical elements constituting spent fuel is feasible is in conflict with the Second Law of thermodynamics. Consequently significant parts of the fission products and actinides remain inevitably in the waste streams of the reprocessing plant.
Furthermore the vitrification concept is (implicitely) based on the questionable assumption that the borosilicate glass will remain stable for hundreds or even 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. How long will the stainless steel containers last in contact with water at elevated temperatures and in the presence of nuclear radiation?
Deep geological repositories are still needed in this waste management option, even more than in case of direct disposal of spent nuclear fuel, because the volumes of highly radioactive waste other than the vitrified fraction are larger. Still no safe final disposal facility for the highly radioactive glass is operational, only paper concepts and experiments exist.

The energy consumption of spent fuel reprocessing is exceedingly high, particularly if the energy investments of the construction and of the cleanup + dismantling of the reprocessing plant would be included. For that reason the energy balance of an LWR system operating in the once-though mode with vitrification might be negative, even including the recycling plutonium and uranium as MOX fuel: the energy requirements to operate such a system from cradle to grave would surpass its useful energy production.

Discharges

Reprocessing is an extremely intricate complex of chemical processes. Each separation step produces inevitably waste streams, containing unwanted nuclides. Even very low concentrations of long-living radionuclides, e.g. I-129, Tc-99 and actinides in ground water or surface water, may be hazardous.

In the reprocessing plant the spent fuel elements are chopped into pieces and the contents are dissolved in boiling nitric acid. The resulting solution contains a host of elements: fission products, uranium, plutonium and higher actinides. The empty cladding hulls of the chopped and leached fuel pins hardly dissolve and are separated from the solution. Not all fuel dissolves either.
A part of the fission products is gaseous (e.g. noble gases) or easily form gaseous compounds and escape from the liquid. Most of that nuclides are difficult to fix, chemically or physically, into solid materials or containers, notably tritium (mainly as HTO), carbon-14 (mainly as $^{14}CO2$), iodine-129 (various compounds, e.g. as $H^{129}I$ or $^{129}I2$) and the noble gases such as krypton-85. The nuclides are discharged into the air or sea. In addition a significant fracion of the soluble nuclides are discharged with the liquid effluents, notably Sr-90, Tc-99, Ru-106 and Cs-137.

Table 2
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.

	NEA 1980 TBq/GW(e).a	Pigford 1973 TBq/GW(e).a	NCRP TBq/GW(e).a	
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 3
Discharge limits of reprocessing plants to the sea in 2000.
n.a. = not available. Source: [OSPAR 2002] Q236

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

Table 4
Discharge limits of reprocessing plants at La Hague.
n.a. = not available. Source: [Malherbe 1991] Q17

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

Data on radioactive discharges by reprocessing plants, and by nuclear power stations as well, are scarce in the open literature.

Lower discharges than the permitted limits do not necessarily mean a better retainment of the radionuclides, but might be caused by a lower throughput of the plant, or by reprocessing spent fuel of lower burnup.

Entropy generation by reprocessing

Reprocessing of spent fuel is an extremely costly and polluting process. Reprocessing greatly enhances the health hazards posed by nuclear power, because many pathways are created along which the radionuclides from the spent fuel can enter the human environment. Reprocessing also raises severe security problems. because fissile materials are separated from the fission products and become accessible.

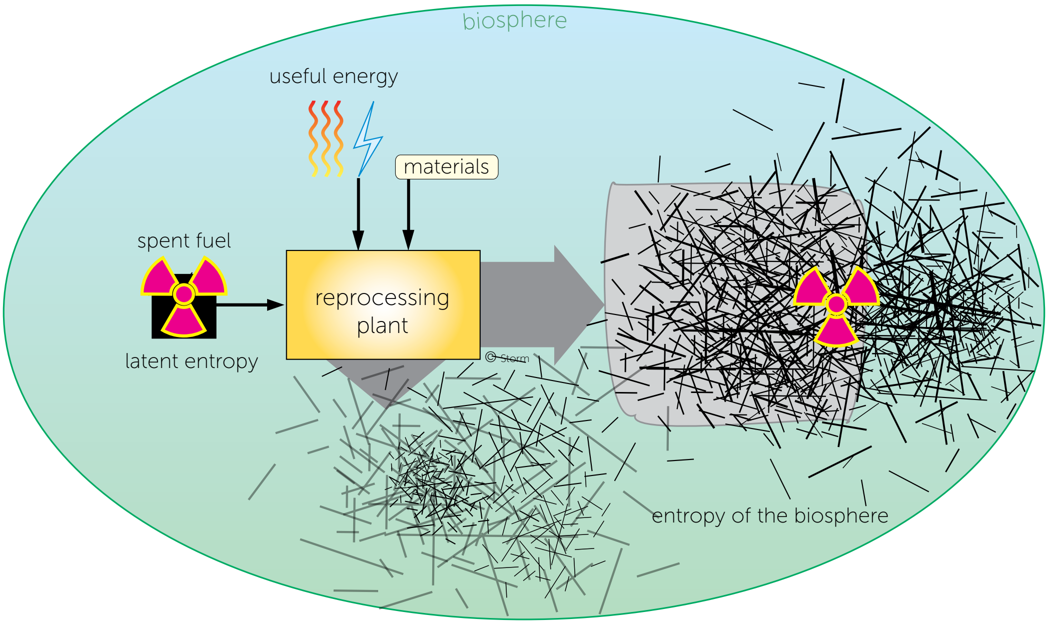

Figure 7

Symbolic representation of the entropy production by reprocessing of spent fuel. In spent fuel is a large amount of latent and delayed entropy confined to the volume of the spent fuel elements. In the reprocessing plant a substantial part of the content of the spent fuel is released into the environment and its latent entropy becomes an acute entropy increase of the biosphere. The other part is distributed over large volumes of originally non-radioactive materials, greatly increasing the entropy of that systems.

In the reprocessing sequence the contents of the spent fuel are distributed over large volumes of non-radioactive materials (see figure 6), greatly increasing the entropy of the spent fuel contents. A signicant part of the radionuclides are discharged into the biosphere via aerosols, gaseous effluents and liquid effluents, causing the conversion of latent entropy into entropy of the biosphere.

New entropy is generated, according to the Second Law, by the consumption of energy and ordered materials in the separation processes. Entropy is also generated by the construction of the reprocessing plant.

In addition to the above mentioned entropy increases, reprocessing generates another form of latent entropy present in the construction materials that became contaminated during the operation of the plant by all kinds of radionuclides from the spent fuel. At issue is to prevent this latent entropy becoming entropy of the biosphere as a result of decommissioning & dismantling of the reprocessing plant itself.

The total quantity of radioactivity does not change by processes in the reprocessing plant, only the extent of dispersion changes.

Reprocessing offers no solution of radioactive waste problems, on the contrary, as is explained in the previous sections on P&T and vitrification. 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, worsening the waste problems beyond control.

The best way to handle spent fuel might be keeping the spent fuel elements intact, - in the fuel elements fissile and other radionuclides are compacted in the smallest possible volume -, to pack them in very durable containers and to dispose of in a safe geological repository (see report m32 *Geologic repositories*). Direct disposal poses the least risks and consumes the least materials and energy, prevents latent entropy in the spent fuel becoming entropy of the biosphere and prevents entropy generation by construction, operation and dismantling.
A number of countries, among other USA, Sweden, Finland 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 needed to achieve reuse of spent fuel are accounted for. The Second Law is relentless.

Concluding notes

Reprocessing of spent fuel is a polluting process, discharging large amounts of radioactive materials into the environment.

Reprocessing increases the risks of severe nuclear accidents, causing dispersion of very large amounts of radioactive materials, possibly more than Chernobyl and Fukushima. Massive amounts of spent fuel are present in reprocessing plants, to be counted in thousands of nuclear bomb equivalents.
Reprocessing increases also the risk of nuclear terrorism.

Reprocessing of spent fuel is an energy-intensive process in itself. Decommissioning and dismantling of a reprocessing plant will require massive investments of energy, materials and human effort. For that reason application of proposed nuclear technologies dependent of reprocessing would result in a negative energy balance of nuclear energy generation.

Separation of any mixture of different chemical species into fractions never goes to incompletion, due to Second Law phenomena. Incomplete separation implies also incomplete purification, so 100% pure materials with 100% predictable properties and behaviour cannot be produced.

All materials are subject to spontaneous degrading processes (ageing), due to Second Law phenomena.

Based on the above observations can be concluded that the following concepts are inherently infeasible:

- uranium-plutonium breeder cycle
- thorium-uranium breeder cycle
- partioning & transmutation, as presented by the nuclear industry.

In addition: inherently safe nuclear power is inherently impossible.

Vitrification of high-level radioactive waste results in an increase of the volumes of radioactive waste that has to be disposed of in geologic repositories, instead of a decrease, as stated by the nuclear industry.

By reprocessing large fractions of the latent entropy confined in spent fuel elements are released as an irreversible entropy increase of the biosphere, in addition to the entropy generation by the processes themselves.

References

Q16
NRC 1996
Rasmussen N C (chair) et al.,
Nuclear Wastes. Technologies for separations and transmutation,
National Research Council, NRC
1996 Washington DC: National Academy Press, 1996.

Q17
Malherbe 1991
Malherbe J,
Nuclear science and technology. Management of radioactive waste from reprocessing including disposal aspects,
Commission of European Community, Directorate-General Science, Research & Development,
EUR 13116 EN
cataogue nr CD-NA-13116-EN-C
Brussels-Luxembourg, 1991,
‹CDNA123116ENC_001.pdf›
http://bookshop.europa.eu/

Q50
Bergelson et al. 2002
Bergelson BR, Gerasimov AS, Kiselev GV & Tikhomirov VG,
Have fast reactors lost the Midas touch?
WNANuclear Engineering International, March 2002, pp. 42-43.

Q58
UNIPEDE/CEC 1981
UNIPEDE/CEC Breeder Reactor Study Group,
Role of breeder reactor system in the European Community,
International Union of the Electric Power Producers and Distributors (UNIPEDE),
Fast Reactor Coordinating Committee (FRCC) and Commission of the European Community (CEC),
Published by ENEL, Roma, September 1981.

Q75
NEA 1980
Radiological significance and management of Tritium, Carbon-14, Krypton-85, Iodine-129, arising from the nuclear fuel cycle,
Report by the NEA Group of Experts
Nuclear Energy Agency (NEA),
OECD, Paris, April 1980.

Q112
Pigford et al. 1973
Pigford Th J, Keaton MJ & Mann BJ,
Fuel cycles for electrical power generation,
prepared for the Standards Research Branch, Office of Research and Monitoring, Environmental Protection Agency,
EPA No 68-01-0561,
Teknekron Report No EEED-101,
Teknekron Inc., California, January 1973.

Q214
Cohen 1977
Cohen BL,
The disposal of radioactive wastes from fission reactors,
Scientific American, vol 236 no 6, June 1977
www.nature.com/scientificamerican/journal/v236/n6/pdf/scientificamerican0677-21.pdf
retrieved 26 Nov 2014

Q236
OSPAR 2002
Liquid discharges from nuclear installations in 2000,
Radioactive Substances Series
OSPAR Commission 2002,
ISBN 0 946956 91 X
‹p00158_Liquid Discharges from Nuclear Installations, 2000.pdf›
www.ospar.org/v_publications/
retrieved 21 November 2013

Q239
Forsey & Dickson 1987
Forsey CD & Dickson RM,
Providing a commercial service for reprocessed uranium,
Nuclear Engineering International, February 1987, pp 28-33.

Q240
Fischer 1986
Fischer U,
Mehrfache Rückführung von Plutonium in thermischen Reactoren,
Atomwirtschaft, November 1986, pp 548-553.

Q241
Roepenack et al. 1987
Roepenack H, Schneider VW & Wittmann K,
Achieving good experience with MOX co-conversion,
Nuclear Engineering International, February 1987, pp 37-38.

Q242
Hulst & Mostert 1979
Hulst PJ vander & Mostert P,
Proliferatiegevaar en kernenergie,
Energiespectrum, februari 1979, pp 38-50 (in Dutch).

Q243
Ronen et al. 2000
Ronen Y, Aboudy M & Regev D,
A novel method for energy production using Am-242m as reactor fuel,
Nuclear Technology, Vol 129, March 2000, pp 407 ff.

Q244
WNA-du 2016
Uranium and depleted uranium,
update September 2016
http://www.world-nuclear.org/information-library/nuclear-fuel-cycle/uranium-resources/uranium-and-depleted-uranium.aspx
retrieved 13 Feb 2017

Q245
WNA-13 2014

Military warheads as a source of nuclear fuel,
World Nuclear Association, info paper#13, November 2007,
www.world-nuclear.org/info/inf13.html
update August 2014
www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Uranium-Resources/Military-Warheads-as-a-Source-of-Nuclear-Fuel/
retrieved Nov 2014

Q246
WNA-mox 2016
Mixed Oxide (MOX) Fuel,
World Nuclear Association, updated October 2016
‹ MOX, Mixed Oxide Fuel - World Nuclear Association.pdf ›
http://www.world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/mixed-oxide-fuel-mox.aspx
accessed 3 March 2017

Q247
WNA-pu 2016
Plutonium
World Nuclear Association, updated October 2016,
‹ Plutonium - World Nuclear Association.pdf ›
http://www.world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/plutonium.aspx
accessed 3 March 2017

Q251
NCRP-62 1995
Tritium in the environment,
National Council on radiation Protection and Measurements,
NCRP Report 62,
Washington DC, January 1995,
(reprint of 1st edition 9 March 1979 and 2nd edition 15 May 1989)

Q254
ORNL-TM-2897 1970
Bell MJ,
Heavy element composition of spent power reactor fuel,
ORNL-TM-2897
Oak Ridge National Laboratory, Oak Ridge Tenessee, May 1970.

Q256
NCRP-81 1993
Carbon-14 in the environment,
National Council on Radiation Protection and Measurements,
NCRP Report 81,
Bethesda, MD, 15 May 1985, first reprinting April 30 1993.

Q259
NCRP-44 1975
Krypton-85 in the atmosphere. Acuumulation, biological significance and control technology,
National Council on Radiation Protection and Measurements,
NCRP Report 44,
Washington DC, July 1, 1975.

Q266
NCRP-75 1983
Iodine-129: evaluation of releases from nuclear power generation,
NCRP Report 75
National Council on Radiation Protection and Measurements,
Bethesda, MD, July 1, 1975.

Q280
MIT 2003-2009
Deutch J et al.,
The Future of Nuclear Power. An Interdisciplinary MIT Study,
Massachusetts Institute of Technology, Cambridge MA, USA, 2003
ISBN 0-615-12420-8
http://www.mit.edu/afs/athena/org/n/nuclearpower/pdf

Update 2009 /2015(?)
http://web.mit.edu/nuclearpower/
retrieved Nov 2018

Q302
WNA-Th 2015
Thorium
World Nuclear Association, updated September 2015
http://www.world-nuclear.org/information-library/current-and-future-generation/thorium/aspx
accessed 7 May 2016, 15 Jan 2017

Q339
Barnaby 2005a
Barnaby F,
Factsheet 1 – Security and Nuclear Power
Oxford Research Group, November 2005
www.oxfordresearchgroup.org.uk/publications/briefing_papers

Q340
Barnaby 2005b
Barnaby F,
Factsheet 2 – Effective Safeguards?
Oxford Research Group, November 2005
www.oxfordresearchgroup.org.uk/publications/briefing_papers

Q360
Barnaby & Kemp 2007
Barnaby F & Kemp J, ed.,
Secure Energy? Civil Nuclear Power, Security and Global Warming,
Oxford Research Group, London, March 2007
www.oxfordresearchgroup.org.uk/publications/briefing_papers/
webpage not available, see:
https://www.files.ethz.ch/isn/29589/secureenergy.pdf
retrieved December 2018

Q376
ORNL-5388 1978
Abbott L S, Bartine D E, Burns T J, ed.
Interim assessment of the denatured 233U fuel cycle: feasibility and nonproliferation characteristics,
ORNL-5388
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, December 1978.

‹ ORNL-5388 ›
http://moltensalt.org/references/static/downloads/pdf/ORNL-5388.pdf
retrieved Sept 2015

Q377
ORNL-6952 1999
Forsberg C W & Lewis L C,
Uses for Uranium-233: What should be kept for future needs?
ORNL-6952
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6180,
September 24, 1999.

Q399
MacKay 2009
MacKay DJC,
Sustainable Energy – without the hot air,
UIT Cambridge Ltd, 2009.
free downloadable file: sewtha.pdf
www.inference.phy.cam.ac.uk/
or
www.withouthotair.com/download.html

Q446
NWMO 2008,
Jackson D P & Dormuth K W,
Watching Brief on Reprocessing, Partitioning and Transmutation and Alternative Waste Management Technology – Annual Report 2008,
Nuclear Waste Management Organization, Toronto, Ontario, Canada,
NWMO TR-2008-22, December 2008,
www.nwmo.ca/

Q447
SKB 2010,
Blomgren J, Karlsson F, Pomp S, Aneheim E, Ekberg C, Fermvik A, Skarnemark G, Wallenius J, Zakova J, Grenthe I & Szabo Z,
Partioning and transmutation. Current developments – 2010,
Technical Report TR-10-35,
Svensk Kärnbränslehantering AB (SKB), January 2010,
www.skb.se/upload/publications/pdf/TR-10-35webb.pdf

Q448
CEA 2002
Radiotoxicity of spent fuel,
Clefs CEA no. 46,
Commissariet à l'Energie Atomique, 2002,
http://www.cea.fr/var/storage/static/gb/library/Clefs46/

Q449
ORNL 2011
Michaels G E,
Partitioning and Transmutation: Making Wastes Nonradioactive,
ORNL, text not dated,
http://www.ornl.gov/info/ornlreview/rev26-2/text/radside1.html
retrieved from web April 10, 2011

Q450
DOE-NE 2009
Advanced Fuel Cycle Initiative,
Department of Energy, Office of Nuclear Energy,
date not given, problably 2009, file retrieved from web April 10, 2011,
http://www.ne.doe.gov/AFCI/neAFCI.html

Q451
LPSC 2001
Brissot R, Heuer D, Ler Brun C, Loiseaux J-M, Nifenecker H & Nuttin A,
Nuclear energy with (almost) no radioactive waste?
Laboratoire de Physique Subatomique et de Cosmologie de Grenoble,
http://lpsc.in2p3.fr/gpr/english/NEWNRW/NEWNRW.html

Q539
ORNL 2013
Partitioning and Transmutation: Making Wastes Nonradioactive,
ORNL Review
http://web.ornl.gov/info/ornlreview/rev26-2/text/radside1.html
downloaded August 2013.

Q541
WNA 2012b
Waste Management: Overview
World Nuclear Association.
http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Waste-Management-Overview/
updated December 2012, retrieved Nov 2015

Q542
WNA 2016c
Radioactive Wastes – Myths and Realities
World Nuclear Association.
http://www.world-nuclear.org/information-library/nuclear-fuel-cycle/nuclear-wastes/radioactive-wastes-myths-and-realities.aspx
updated February 2016, retrieved August 2016
(formerly WNA 2012c)

Q552
ISIS 1999
Albright D & Barbour L,
Trouble Tomorrow? Separated Neprunium-237 and Americium,
Chapter 5 of:
Albright D & O'Neill (eds)
The Challenges of Fissile Material Control,
Institute for Science and International Security,
ISBN 0-9669467, Washington, March 1999.
‹New chapter 5.pdf›
www.isis-online/publications/fmct/book/
retrieved 22 September 2013

Q587
KfK 1983
Fischer U & Wiese HW,
Improved and Consistent Determination of the Nuclear Inventory of Spent PWR Fuel on the Basis of Cell-Burnup Methods Using KORIGEN,
KfK 3014 (ORNL-TR-5046),
Kernforschungszentrum Karlsruhe, January 1983.
‹KfK-3014en.pdf›
www.oecd-nea.org/science/wpncs/ADSNF/reports/ICE/

retrieved 10 December 2013

Q590
Schneider 2007
Schneider M,
The permanent Nth country experiment. Nuclear weapons proliferation in a rapidly changing world,
Paris, 24 March 2007,
commissioned by The Greens|European Free Alliance.
‹07-03-18_MycleNthCountryExperiment-2.pdf›
www.cornnet.nl/~akmalten/
retrieved 14 February 2014

Q591
Frank 1967
Frank WJ,
Summary report of the Nth country experiment,
Lawrence Radiation Laboratory, University of California, Livermore
March 1967.
‹nth-country.pdf›
www2.gwu.edu/~nsarchiv/news/20030701/
retrieved 14 February 2014

Q593
Glaser 2005
Glaser A,
Neutronics calculations relevant to the conversion of research reactors to low-enriched fuel,
Dissertation, Technische Universität Darmstadt, 27 April 2005.
‹aglaser_thesisrev.pdf›
http://tuprints.ulb.tu-darmstadt.de/566/1/
retrieved 14 February 2014

Q594
Alvarez 2012
Alvarez R,
Managing the uranium-233 stockpile of the United States. Questionable conditions of taking inventory, storing, and disposing of a nuclear explosive material,
Institute for Policy Studies, August 2012.
‹Bob-Alvarez-Managing-Uranium.pdf›
www.ips-dc.org/reports/managing_uranium

Q599
O'Connor et al. 2003
O'Connor GJ, Bowden RL & Thorne PR,
Burn-up Credit Criticality Benchmark. Phase IV-A: Reactivity prediction calculations for infinite arrays of PWR MOX fuel pin cells,
NEA/NSC/DOC(2003)3
Nuclear Energy Agency OECD/NEA, April 2003.
‹NSCDOC(2003)3-BUC-4a.pdf›
www.oecd-nea.org/science/wpncs/Publications/BUC/
retrieved 15 Febr 2014

Q617
PSR-IEER 2009
Makhijani A & Boyd M,
Thorium fuel: no panacea for nuclear power,
Factsheet by Physicians for Social Responsibility (PSR) and Institute for Energy and Environmental Research (IEER).
January 2009, updated July 2009.
‹thorium2009factsheet.pdf›
http://ieer.org/wp/wp-content/uploads/2012/04/thorium2009factsheet.pdf

Q667
ENS 2016
Repropcessing plants, world-wide,
European Nuclear Society,
https://www.euronuclear.org/info/encyclopedia/r/reprocessing-plants-ww.htm
retrieved 9 Jan 2016

Q668
nucl-reproc-wiki 2016
Nuclear reprocessing
Wikipedia, 5 Jan 2016
https://en.wikipedia.org/wiki/Nuclear_reprocessing
retrieved 12 Jan 2016

Q693
wiki-msr 2017
Molten Salt Reactor
Wikipedia, last modified 27 February 2017
https://en.wikipedia.org/wiki/Molten_salt_reactor
accessed 4 March 2017

Q734
Pistner et al. 2015
Pistner C, Englert M, Schmidt G, Kirchner G,
Partitoning & Transmutation. Solution for nuclear waste?
1st NURIS Conference, Vienna, 17-17 April 2015
‹ Pistner_Partioning-and-Transmutation.pdf ›
http://nuris.org/wp-content/uploads/2015/04/Pistner_Partitioning-and-Transmutation.pdf
accessed 23 Feb 2017

Q738
WNA-reproc 2016
Processing of Used Nuclear Fuel,
World Nuclear Association, updated November 2016,
‹ Processing of Used Nuclear Fuel - World Nuclear Association.pdf ›
http://www.world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/processing-of-used-nuclear-fuel.aspx
accessed 3 March 2017

Q739
WNA-msr 2016
Molten Salt Reactors,
World Nuclear Assocation, updated 30 September 2016,
‹ Molten Salt Reactors - World Nuclear Assocation.pdf ›
http://www.world-nuclear.org/information-library/current-and-future-generation/molten-salt-reactors.aspx
accessed 4 March 2017

Q740
WNA-ane 2016
Accelerator-driven Nuclear Energy,
World Nuclear Assocation, updated March 2016,
‹ Accelerator-driven Nuclear Energy|Accelerator Driven Systems|Transmutation - World Nuclear Assocation.pdf ›
http://www.world-nuclear.org/information-library/current-and-future-generation/accelerator-driven-nuclear-energy.aspx
accessed 4 March 2017

Q741
wiki-mox 2017
MOX fuel,
Wikipedia, 25 February 2017
‹ MOX fuel - wikipedia.pdf ›
https://en.wikipedia.org/wiki/MOX_fuel
accessed 4 March 2017

Q742
IAEA-p&t 2017
Nuclear Fuel Cycle and Materials. Partitioning and transmutation,
https://www.iaea.org/OurWork/ST/NE/NEFW/Technical-Areas/NFC/advanced-fuel-cycles-01-partitioning and transmutation.html
accessed 23 February 2017

Q744
CNRS/EDP 2017
‹ Radioactivity : Spent fuel composition.pdf ›
http://www.radioactivity.eu.com/site/pages/Spent_Fuel_Composition.htm
accessed 14 March 2017