Partitioning & transmutation

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Note

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

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Summary

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

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

The concept of partitioning and transmutation involves a complex cycle of advanced technical processes. At this moment the P&T cycle exists only in cyberspace.
Partitioning is the separation of spent nuclear fuel into different fractions: uranium, plutonium, other actinides, long-lived fission products and the remaining content of spent fuel.
Transmutation occurs in a special nuclear reactor, where the long-lived radionuclides and actinides are converted into short-lived radionuclides, or stable nuclides, by neutron irradiation.

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, a few 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.

Introduction

Concept

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, and the activity of the actinides with a factor of ten thousand. After that period, the radioactivity decreases very slowly.

Figure 1
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. Sources: [Bell 1973] Q264, [Hollocher 1975] Q262, [JPL-77-69 1977] Q263, [Charpak & Garwin 2002] Q300.

Theoretically it is possible to convert long-lived radionuclides, not all, 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 from spent nuclear fuel 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 costly 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). 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

The P&T system

A partitioning & transmutation system consists of a transmutation cycle and a waste management system. The cycle consists of three components (see Figure 2):

- 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.

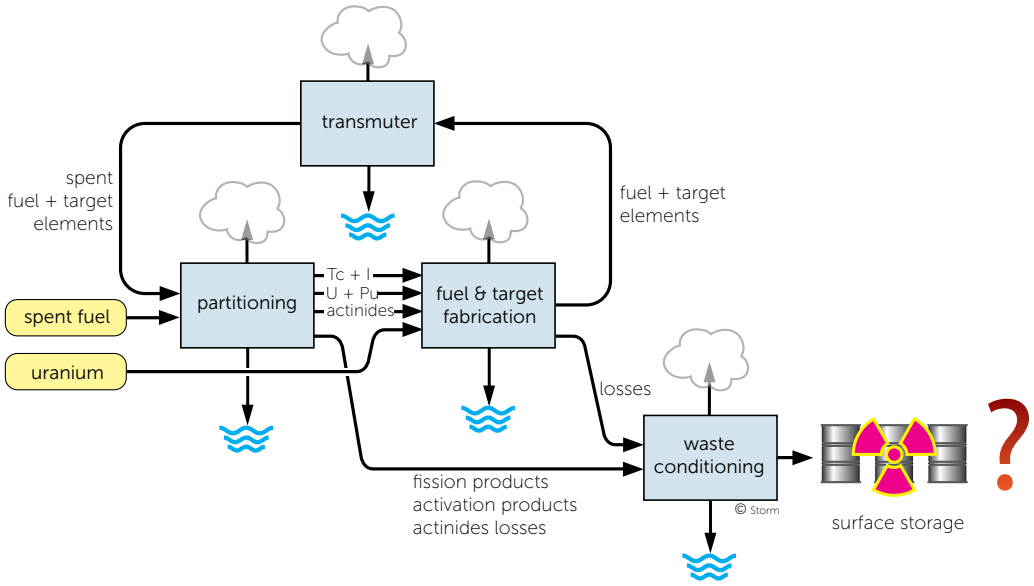

Figure 2

Flows of radioactive materials in the proposed concept of a partitioning & transmutation system. The radioactive wastes from the cycle are to be conditioned and stored in storage facilities at the surface or shallow burial, according to the concept. The querie symbolises the uncertainty about this storage method. 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. All components of the P&T cycle discharge radioactive materials into the biosphere.

Transmutation

Two nuclear processes are important in a P&T system:

- transmutation by ineutron capture (activation)
- fission of actinides

Both processes result in the generation of nuclides of elements other than the original ones.
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, and is undesired
- An unstable nucleus transforms into another unstable nucleus of the same element with a shorter half-life than its parent.

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 (partition).
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.

Partitioning

Spent fuel contains a mixture of many dozens of different kinds of atoms (chemical elements), often with different isotopes. This complicated mixture has to be sorted into several fractions in a process called partitioning, a crucial step in the P&T cycle.

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.

Partitioning

Outline

Spent fuel contains a mixture of many dozens of different kinds of atoms (chemical elements), often with different isotopes. This 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 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.

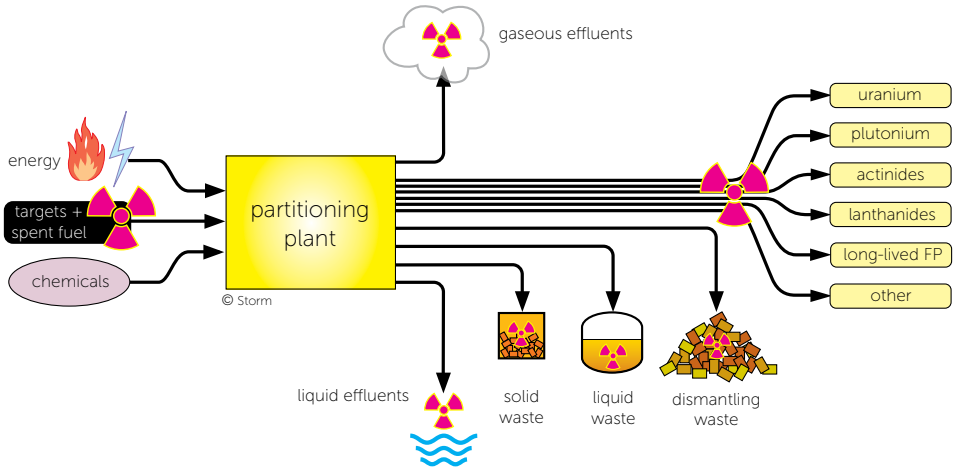

Figure 3
Outline of the partitioning of spent nuclear fuel and target elements of a transmutation cycle. For details see text.

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.

Partitioning would meet the same kind of problems as conventional reprocessing, but to a much greater extent, because partitioning of spent fuel is a much more demanding process than conventional reprocessing. The separation processes are more complicated for several reasons:

- Spent fuel and target elements from the transmuter have to be partitioned into more pure fractions than in conventional reprocessing.
- Elements with similar chemical properties have to be separated, for example lanthanides from actinides.
- Due to the high specific radioactivity of the liquid mixture from which the various fractions are to be extracted radiolysis of the separation chemicals require short contact times between the organic and aquous phase, causing less complete separation and consequently higher process losses. This effect increases with each cycle caused by the increasing radioactivity of the fuel elements and target elements with each cycle, so the process losses will rise with each cycle.

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 advanced thermal neutron reactors, mostly LWRs, not more than about 0.5% 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 see reports m01 *Uranium-plutonium breeder systems* and m38 *Nuclear power and the Second Law*.

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.

U and Pu in closed-cycle reactors

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.

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.

Feasibility of the P&T system

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 report m20 *Reprocessing of spent nuclear fuel*.

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 sub-critical 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 report m38 *Nuclear power and the Second Law*.

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 report m20 *Reprocessing of spent nuclear fuel*, 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] Q127, [RAND 1981] Q126. 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 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, according to [UCS 2007] Q421.

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.

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.

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 into the waste streams.
- It is not possible to produce materials which are 100% pure and with 100% predictable properties.
- 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 may be 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.

More details are discussed in report m38 *Nuclear power and the Second Law*.

Health hazards

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 much larger than 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 severe accidents, and consequently would inrease the health risks of nuclear power.

Potential, direct health hazards

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 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 a mix of radionuclides.
- 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.

Additional health hazards posed by P&T

Health hazards would increase greatly by the introduction of P&T, compared to the hazards caused by the current 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 of materials
- more radioactivity in chemical and physical mobile state during periods measured in centuries
- more discharges of human-made radioactivity into the environment due to the inherent process losses of the partitioning plant and the inherent imperfections of the other systems in the P&T cycle
- greater chances of severe accidents involving very large amounts of radioactive materials
- more risks posed by unpredictable human behaviour
- 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.

Even without materialisation of a P&T system the concept alone introduces 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 day of postponement enhances the chances of accidents involving large amounts radioactive waste. Each day of postponement the unintended but unavoidable discharges of radioactive materials from temporary waste storage facilities into the environment are continuing.

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