Uranium-plutonium breeder systems

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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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Introduction

The only fissile nuclide in nature is uranium-235, accounting for 0.7% of the atoms in natural uranium. The remaining 99.3% consists of uranium-238 atoms and traces of uranium-234 atoms, which are not fissile.
Nuclear reactors generate energy through fission, the process by which a uranium-235 nucleus splits into two or three smaller nuclei. During fission energy is released as radiation and heat. The heat is used to raise steam, which drives a turbine coupled to an electricity generator.

When a uranium nucleus fissiones it produces two or three free neutrons. A part of the new neutrons is absorbed by fissile nuclei causing new fissions, thus maintaining a fission chain reaction. The other neutrons are captured by the nuclei of uranium-238, coolant and construction materials.
If a neutron is captured by an atomic nucleus of the coolant or construction, a new nucleus with one neutron more emerges. Often the new nucleus is instable, and radioactive, hence the name 'activation reaction' of neutron capture reactions. In this way the materials of a nuclear reactor, its coolant and its surrounding appendages become radioactive, the more so the longer the reactor operates and the higher its power.
If a neutron is captured by a uranium-238 nucleus, a uranium-239 atom is formed. By radioactive decay a uranium-239 nucleus transmutes into the nucleus of plutonium-239, which is fissile.

Neutrons set free by a fissioning uramium-235 or plutonium-239 have high energy and are moving extremely fast. These fast neutrons do not efficiently cause fission. In thermal-neutron reactors the fast neutrons are slowed down by a moderator – light water, graphite or, in some cases, heavy water – to energies in thermal equilibrium with the atoms and molecules in the reactor, hence the name 'thermal neutrons'. All power reactors currently operating (except one) are thermal-neutron reactors, 88% of them being light-water reactors (LWR).
Little more than about 0.5% of the atoms in the natural uranium leaving the mine can be fissioned in thermal-neutron reactors, even the best currently operating reactors attain a uranium utilization of about 0.5%. This implies that 99.5% of the uranium atoms extracted from the earth's crust are disposed of, unfissioned, in the nuclear waste.

In an operating nuclear reactor the abundant and 'fertile' uranium-238 partially is converted by neutron capture into plutonium-239, which is even better fissile than uranium-235. In a fast reactor the neutrons are not slowed down, by using non-moderating coolants, such as liquid sodium. Fast neutrons, though not as good at causing fission, are readily captured by uranium-238 atoms. Theoretically, in special designs of a fast reactor, more plutonium atoms could be formed from U-238 than are fissioned. Therefore these reactors are dubbed 'breeders' and often 'fast breeders', that does not mean the breeding process goes fast. Besides, not every fast reactor is a breeder.
This concept theoretically would make possible to fission 30-60% of the atoms in natural uranium in a breeder reactor, via conversion into plutonium. That would mean 50 to 100 times the fissioned amount in conventional (thermal neutron) reactors.

The high fissionable fraction of natural uranium theoretically attainable by the breeder is the source of the old nuclear promises from the 1950s: 'all nuclear society', 'too cheap to meter' and 'burning the rocks'. Today these unproven figures still give rise to promises of untold quantities of cheap, clean nuclear energy for all mankind for the next centuries.
However, there are serious obstacles on the road to the materialization of these technical dreams:

- technical unfeasibility of the breeder system
- slow rate of expansion of a plutonium economy due to limited availability of fissile material, and the long doubling time of the breeding cycle
- uncontrollable and high risk of plutonium terrorism and the proliferation of nuclear technology.

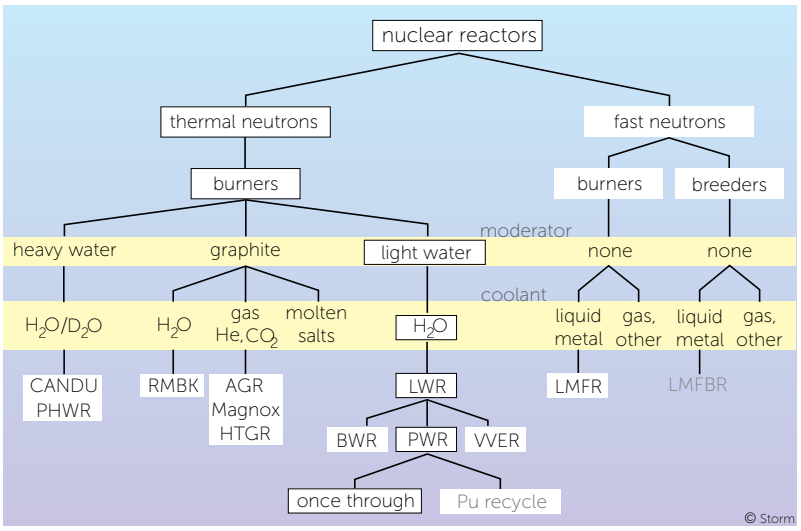

Figure 1

Decision tree to define the reference nuclear reactor. Only that types of power reactors are mentioned at the bottom of the diagram that are actually operating or have been operating. The LMFBR is not operational. The LWR has been chosen as the reference reactor type in this study. The differences between the Boiling Water Reactor BWR, Pressurized Water Reactor PWR and the Russian VVER (also a PWR) with respect to the energy analysis are minor.

Burner reactors

All power reactors (except one) currently operating are burner reactors, based on fission with thermal (slow) neutrons. The three main classes are (see also Figure 1):

- light-water reactors LWR: Pressurized Water Reactor PWR and Boiling Water Reactor BWR,
- graphite-moderated reactors and gas-cooled reactors, e.g. Magnox and AGR
- heavy-water moderated reactors, e.g. CANDU.

At present 88% of the power reactors of the world are LWRs (see Figure 2).

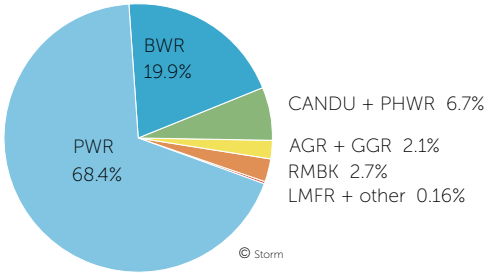

Figure 2

Distribution of the world nuclear power reactors in 2007, according to power output. The section PWR includes the Russian version (PWR-VVER). 88.4% of the world nuclear capacity is supplied by light-water reactors (LWR) and 11.6% by other types. Source: European Nuclear Society [www.euronuclear.org/info/].

The reference reactor of this study, which may serve as a model for the newest currently operating power reactors, achieves a lifetime uranium utilization of about 0.5%. This implies that 5 grams of each kilogram natural uranium as delivered by the mine, actually are fissioned. The remaining 995 grams leave the nuclear energy system as depleted uranium and highly radioactive spent fuel.

Advanced reactors, such as the so-called Generation III reactors and the Pebble Bed Reactor PBMR, all are burner reactors and may reach a uranium utilization of somewhat higher than 0.6%, but still lower than than 0.7%. One of the first Generation-III reactors, an EPR (European Pressurized Reactor), is being built at Olkiluoto in Finland. As none of the advanced reactors are operating yet, no operational data are available.

Breeder cycle

What is called a 'breeder' is not just a reactor type or a stand-alone system. To exploit the promised potential of natural uranium, a complex breeder cycle is prerequisite.
The cycle (see Figure 3) comprises three components: the breeder reactor, a reprocessing plant and a fuel fabrication plant. All three have to operate flawlessly and continuously and have to be finely tuned to each other. If one component fails, the whole breeding system fails. If the system losses are higher than the breeding gain, the system fails as breeder. Then you may run a fast reactor, but you do not have a breeder.
The light-water reactor (LWR) system can be operated in an open 'cycle' (once-through mode) and has no such dependency.

Breeder reactor

The active core of the breeder reactor is surrounded by a blanket of uranium-238, the fertile material. In the core plutonium is fissioned, producing heat and neutrons to sustain the fission process and to 'breed'. In the blanket U-238 atoms are converted into plutonium atoms by capture of neutrons from the core. In theory it is possible to breed more fissile material (plutonium) from the U-238 in the blanket than is fissioned in the core. So, the breeder can be taken as an indirect way of fissioning the non-fissile uranium-238.

Most designs are based on the liquid-metal cooled fast-neutron reactor (LMFR), but other concepts of U-Pu breeder reactors have been conceived. The sodium-cooled fast-neutron reactor reached the prototype stage with the French Superphénix. This paper confines to the LMFBR (liquid metal fast breeder reactor), but the principal difficulties apply to all other types of breeders, e.g. Generation IV reactors.

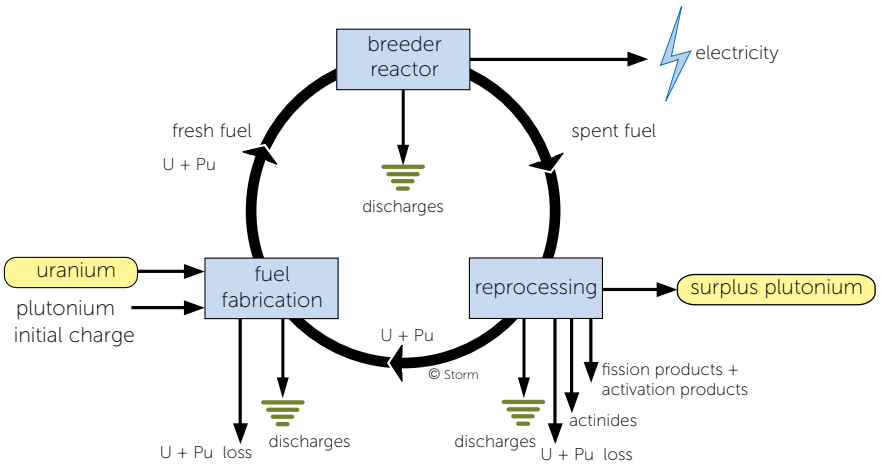

Figure 3
General outline of the breeder system in steady state. By repeatedly recycling spent fuel, it would be theoretically possible to fission the main part of natural uranium The cycle represents the mass flows of uranium and the nuclides originating from the uranium by nuclear processes in the reactor: fission, neutron capture and radioactive decay.

Reprocessing

The second step of the breeding cycle is the reprocessing of the spent fuel and irradiated blanket elements, to separate the plutonium and remaining uranium from the fission products and the unusable and dangerous actinides (Np, Am, Cm and higher nuclides).

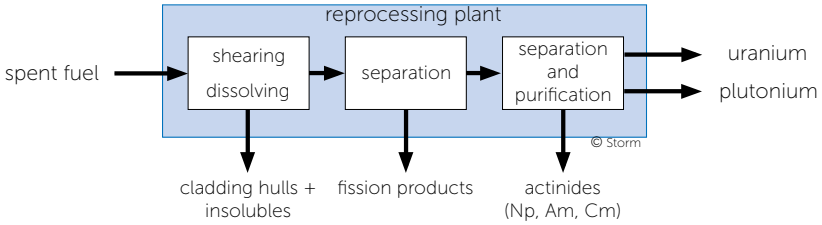

Figure 4
General outline of the reprocessing of spent fuel

The partitioning process comprises a number of chemical equilibrium reactions. A basic consequence of chemical equilibria is that the extraction of one chemical species from a mix of a number of chemical species never goes to completion. This implies an incomplete separation: the waste streams always will contain small quantities of the wanted for species, in case uranium and plutonium, and the product stream of U and Pu will contain small quantities of the 'waste nuclides': fission products and unwanted actinides. The contamination of the products U and Pu with other nuclides causes a higher radioactivity and a less predictable behaviour in the reactor of the recovered uranium and plutonium.
After each breeding cycle the radioactivity of the fuel increases by cumulation of highly radioactive nuclides which are not removed from the fuel in the reprocessing [NRC 1996] Q16. The high burnup of the fuel – typically 100 GW(th).day/Mg U – needed for economic reasons enhances this effect.

Fuel fabrication

The third step is the fuel fabrication facility, to make new fuel elements from the recycled plutonium and uranium from the reprocessing plant, replenished with 'fresh' uranium (depleted uranium or natural uranium). The 'fresh' fuel could then be reinserted into the reactor.

The breeding system

A breeding ratio higher than 1 means: for every fissile atom fissioned in the reactor, more than 1 new fissile atom is created in the blanket from non-fissile atoms by neutron irradiation.
If the system works as designed, the cycle would produce during its operational life span a plutonium gain, large enough to start up two (or a little more) new breeders: one to replace the closed down unit, and one (or more) additional breeder(s).
The initial plutonium charge to start up the breeder reactor here is set at 3 Mg fissile Pu for a 1 GW(e) FBR.

Main parameters

A few important technical parameters of the breeder system are summarized in Table 1.
The first three are reactor parameters, the latter two are set by the two other components of the cycle: reprocessing and fuel fabrication. Some technical hurdles of the breeder cycle are discussed below.

Table 1

parameter	should be:
• initial inventory of plutonium in Mg/GW	as low as possible
• breeding ratio	as high as possible
• full-power operating time of the reactor	as long as possible
• out-of-core time of the plutonium	as short as possible
• plutonium losses in the cycle	as low as possible

During the build up of an energy system based on breeders, the breeding factor of the whole cycle should be larger than one, in order to launch two new breeders each time an old one is retired at the end of its operational life and is closed down: one to replace the closed-down unit and the other to extend the energy system. In this way the number of breeders would grow by time.
The doubling time of the breeder system – the time one reactor needs to produce enough net plutonium to start up two new breeder reactors – should be as short as possible.

Plutonium recycling

The composition of plutonium leaving an LWR is summarized in Table 2. The composition of the plutonium produced by a breeder with a high fuel and blanket burnup will be slightly different: the share of the higher-mass isotopes (Pu-241, Pu-242 and heavier) will be higher. The basic problems pointed out below remain similar, if not worse.

Table 2
Composition of the plutonium generated in an LWR.

Isotope	percentage	fissile?	half-life(years)	decay mode
Pu-238	2	no	87.8	alpha, gamma
Pu-239	59	yes	24390	alpha, gamma, spontaneous fission
Pu-240	24	no	6540	alpha, gamma
Pu-241	11	yes	15	beta, gamma, spontaneous fission
Pu-242	4	no	387000	alpha, gamma

Reactor-grade plutonium contains roughly 70% fissile nuclides. To load a breeder reactor with 3 Mg fissile plutonium, 4.3 Mg reactor-grade plutonium has to be inserted into its core.

Pu-238 decays to U-234 and Pu-241 to Am-241, with relatively short half-lifes. The longer the out-of-core time of the spent fuel, the more Pu-238 and Pu-241 have decayed. The total amount of plutonium decreases with time, as does the fissile fraction of the remaining plutonium. The decay products, U-234 respectively Am-241, have adverse properties in a reactor, apart from their high gamma emission.
After reprocessing the storage time of the plutonium before reuse should be kept as short as possible, to prevent Am-241 building up.

After reinsertion into the reactor, the recycled plutonium partially fissions, partially remains unaltered and partially captures one or more neutrons and transmutes to a heavier plutonium isotopes or to a trans-plutonium actinides. The higher the burnup of the fuel, the stronger this effect. In this way a part of a given

mass of plutonium gets lost from the fissile inventory. Each recycle this effect worsens. After a number of cycles a given mass of plutonium has been converted in this way into non-fissile nuclides and/or nuclides that are removed in the reprocessing plant.

Summarizing, the plutonium cycle has three intrinsic mechanisms causing loss of fissile plutonium:
- decay of Pu-238 and Pu-241
- transmutation by neutron capture into non-fissile nuclides.
- losses in the separation processes of the reprocessing plant.

At one hand a high burnup of the fuel in a breeder reactor is desirable to breed as much as possible plutonium in the blanket before refueling the reactor, but at the other hand a low burnup yields a more favourable isotopic composition of the newly bred plutonium. A lower burnup means a shorter stay time of the blanket and fuel elements in the reactor and the need for more frequent reprocessing of the irradiated material. During reprocessing inevitably a few percents of the plutonium gets lost in the waste stream. For that reason an optimum should be found between two desirable conditions.

Every cycle the separation losses during reprocessing increase, due to, among other:
- higher content of actinides
- materials are more radioactive
- shorter contact time caused by radiolysis, hence lower extraction yield
- stronger neutron radiation caused by spontaneous fission; this phenomenon causes also that the fuel becomes less and less predictable in reactor
- increasing criticallity problems with actinides due to low critical mass.

Due to their high specific activity Pu-238 and Pu-241 are making plutonium difficult to handle.

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.

Technical hurdles of the breeding system

Breeder reactor

During the past six decades several designs of fast-neutron breeder reactors have been proposed. Three main types are:
- the liquid metal-cooled fast reactor,
- the molten salt fast reactor and
- the high-temperature gas-cooled fast reactor.

The first small experimental fast reactors have been tested during the 1940s and 1950s. During the 1950s and 1960s research on breeders started.

Six countries seriously attempted to materialize the promises of the breeder concept and to develop the breeder cycle: USA, UK, France, Germany, Japan and the former Sovietunion. Large-scale development projects of fast breeders ran during the 1970s through 1990s, involving investments of hundreds of billions of dollars. The breeder designs in these six countries all were based on the liquid sodium-cooled fast breeder reactor concept. The breeder programmes of China and India started later and are of modest scale. The liquid metal-cooled fast breeder reactor (LMFBR) is the only breeder concept that reached the demonstration and prototype phases.
- Fast reactors are more difficult to control than thermal-neutron reactors, due to the fast neutron spectrum.
- Under some conditions, e.g. boiling sodium, the fission process is difficult to contain and a power excursion or run-away chain reaction is possible, causing an explosion and/or a core meltdown. A liquid

sodium-cooled fast reactor can be made safe only by engineered safety measures.
- Ageing and embrittlement of the materials of the fuel elements and the reactor construction under the harsh conditions of high temperatures (500-600 °C) and high fluxes of fast neutrons.

The liquid sodium-cooled fast reactor are inherently unsafe. Contact of liquid sosium (leaks) with water and or air may cause severe accidents.
The liquid sodium has to be kept extremely pure, in order to prevent build up of radioactivity from activated corrosion products and impurities and to prevent precipitation of solid chemical reaction products, clogging ducts and coolant passages.
The flow rate of the liquid sodium through the reactor core has to be high, due to the high power density of the core: very large amounts of heat are generated within a small volume.

Process losses

Material losses in the breeder cycle are inevitable: 100% recovery of the U and Pu from the spent fuel is impossible. Part of the uranium and plutonium will be lost in the waste streams which are inherent to all separation processes.

Losses during fuel fabrication may be higher for FBR fuel than LWR fuel, because of the high specific radioactivity of the plutonium and recycled uranium.
Another part of the fertile and fissile material is lost from the breeder cycle by conversion into trans-plutonium actinides (Am, Cm), that are separated from the plutonium in the reprocessing plant. The longer the out-of-pile time of the recycled uranium and plutonium, the more fissile material will be lost by decay into higher actinides.

Reprocessing

Reprocessing of FBR fuel, which would have a high burnup (typically 70-100 GW(th).day/Mg, or even higher) 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 (= amount of neutrons absorbed per unit volume). Higher neutron flux implies higher concentrations of plutonium, of trans-plutonium actinides, of fission products and of radioactive activation products. Spent FBR fuel is a much more 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 produces more radiolyse (products of radiation) in 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 insoluble compounds and noble metal alloys (Ru, Rh, Tc, Mo, Pd), possibly containing plutonium, and the undissolved mixture of plutonium and uranium oxides (MOX) particles remain in the dissolver. The undissolved 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 effectiveness 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 necessitates 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 smaller 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 – preferably less than 1 year (versus LWR fuel cooling times of 3 years or more) – the shearing of the fuel elements releases highly active short-lived gaseous fission products, such as I-131. The effluent release after short cooling times causes two additional problems. First, the higher decontamination factors required on gaseous effluent for reduction to acceptable operational release levels. Secondly, 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 more hazardous 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 according to [UNIPEDE/CEC 1981] Q58. The study of [NRC 1996] Q16 observes exponentially rising reported costs of the reprocessing of LWR fuel during the past decades.
The specific energy consumption of reprocessing may be high, especially when the decommissioning and dismantling costs of the reprocessing facilities are accounted for.

Fuel fabrication

The problems with recycled plutonium and uranium – increasing gamma and alpha activity and neutron radiation by spontaneous fission, caused by higher Am-241 content – worsen with every cycle.
The decay of Pu-241, one of the plutonium isotopes formed in the reactor, to Am-241 demands a high turnover rate and out-of-core stay times as short as possible, to avoid the need for repurifying the plutonium, a costly and energy consuming process.
The technical difficulties of MOX fuel fabrication for LWRs, are valid to a larger extent for the fabrication of fast breeder reactor fuel.
Costs of fuel fabrication in the breeder cycle are estimated at 2-4 times as much as LWR fuel [UNIPEDE/CEC 1981] Q58. The fuel elements have to be fabricated under remote control, because of the high specific radioactivity of the materials to be processed. Little, if any, experience exists in remote operation and maintenance of fuel fabrication facilities and in a full-fledged breeder program numerous high-capacity facilities are needed [NRC 1996] Q16.

Uranium recycling

Recycled uranium, emerging from reprocessed nuclear fuel, has a different composition than natural uranium and contains a number of nuclides that deteriorate its properties as nuclear fuel [Foresey & Dickson 1987] Q239, among other:

- 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 U-236 needs a higher enrichment in U-235 or more fissionable plutonium (Pu-239 and Pu-241) to compensate for it
- traces of fission products – the chemical separation in the reprocessing plant never is 100% perfect – such as ruthenium-106 and technetium-99, which increase the gamma activity of the recycled uranium
- traces of trans-uranium elements, e.g. neptunium and plutonium.

Above problems worsen each time the fuel passes the fuel cycle.

Re-enrichment of recycled uranium would result in a product enriched in the unwanted nuclides U-232 and U-234 and, to a lesser extent, U-236. Due to the neutron-absorbing properties of these uranium isotopes, a higher content of fissile nuclides (U-235 or Pu-239 + Pu- 241) is needed to maintain the fission process in the reactor. Re-enrichment of recycled uranium would have the additional drawback of contaminating the enrichment plant with highly radioactive species, seriously hampering it operation.

Feasibility of the U-Pu breeder system

All three components of the breeder cycle must operate flawlessly and exactly tuned to each other, before any breeding can be achieved. If one component fails, the whole system fails. In fact, none of the three components has ever demonstrated operation as required, let alone the three components together as one integrated continuously operating system.
Sixty years of intensive research in eight countries (USA, UK, France, Germany, former USSR now Russia, Japan, India and China), with investments of tens, if not hundreds of billions of dollars so far have failed to demonstrate that the breeder cycle is technically feasible.
To day three fast-neutron sodium-cooled reactors (LMFR) are more or less operable in the world: Monju in Japan, Beloyarsk-3 in Russia and Phénix in France [NEI, May 2003] Q235. Only the Russian reactor is operating, but it never bred, and it has a history of large and serious accidents. Although designed as breeders, none actually bred. It is not clear whether the French and Japanese reactors, out of operation for years, will ever be restarted.
Problems of the breeder system are discussed in detail by, among others, [Lidsky & Miller 1998] Q301. The authors concluded that the breeder system is not feasible, not only due to the technical hurdles, but also because the system cannot meet the requirements of safety, proliferation and economy.
The [MIT 2003-2009] Q280 study The Future of Nuclear Power, does not expect breeders (in effect breeder cycle) to come into operation during the next three decades.

Today only Japan, India, China and Russia have operational breeder programs, may be on a modest scale. During the past decade no significant progress in the breeder development has been reported, let alone the major breakthroughs required to make the breeder system technically feasible.
No (new) research projects aimed at the development of the breeder cycle are reported in the USA, the UK, France and Germany, as far as known. The latter four countries effectiviely shut down their

LMFR as transmuter

Within the nuclear industry proposals are circulating about using fast reactors (LMFR) as plutonium burners and as actinide transmuters, to destroy plutonium inherited from the past and to shorten the radioactive

decay of nuclear wastes which contains actinides. As it turns out, fast reactors are hardly more efficient than LWRs for the transmutation of actinides [Bergelson et al. 2002] Q50.
Moreover the transmutation of long-living nuclides into short-living nuclides is a purely hypothetical concept, existing only on paper. At best a small part of the long-living nuclides could be transmuted in a favourable way, and even that part would take centuries to attain a reduction to less than 10% of the original quantity. [NRC 1996] Q16.

New names, no new concepts

Presently the nuclear industry avoids the term 'breeder' or 'LMFBR' and uses preferably the terms 'fast reactor' or 'Generation IV reactor' or 'closed-cycle reactor'. When speaking about these reactor types the nuclear world usually has fast U-Pu breeder reactors (LMFBR) in mind. From a publicity point of view this change of name has an understandable reason, because the concept proved to be technically unfeasible and consequently 'breeder' and 'LMFBR' connote failed concepts.

The failure of the breeder concept is not caused by protests of environmental activists or by actions of leftist politicians, nor for economic reasons, as the nuclear industry asserts, but is caused by fundamental technical limitations. Implicitely the nuclear world assumes the availability of 100% perfect materials and 100% complete separation processes. None of these two conditions are possible, as follows from the Second Law of thermodynamics. By virtue of this law can be argued beforehand that the breeder cycle likely will not work.

Logistic hurdles

During the building up of an energy system based on breeders the doubling time of the system should be as short as possible. Based on the LMFBR technology represented by the design characteristics of the experimental breeder reactors of the 1980s, the breeder system would have a doubling time of about 87 years [UNIPEDE/CEC 1981] Q58.

Plutonium availability

Even if the breeder system starts working according to the textbook from now on, a logistic problem is limiting the set up of a large-scale breeder-based energy system: the plutonium availability. The first breeders are to be fueled with plutonium from LWRs. The global stockpile of reactor-grade plutonium, extracted from civil spent fuel, is estimated at more than 260 metric tonnes as of 31 December 2011 [IPFM 2011] Q513 and is stored in a number of facilities around the world. In addition 150-200 Mg weapons-grade plutonium is in stock, according [WNA-pu 2016] Q247. Assuming an initial charge of 3 Mg of fissile plutonium per 1 GW breeder reactor, corresponding with 4.3 Mg reactor-grade Pu, the global inventory of reactor-grade Pu would be adequate to start up about 56 breeder reactors. It seems not likely that also the weapons-grade plutonium would be used in breeder cycles.

Two scenarios may demonstrate the potential of the breeder system as part of the world energy supply. Both scenarios are based on the following assumptions. It should be noted that each assumption in itself is extremely unrealistic, let alone the combination of the seven assumptions.

- the breeder system works according to the textbook,
- the doubling time of the system is 40 years (a survey based on the state of technology in the early 1980s estimated a doubling time of 87 years [UNIPEDE/CEC 1981] Q58,

- in 2020 the construction programme of the maximum number of breeder systems starts
- the number of breeders is limited only by the amount of available plutonium,
- in 2030 all planned breeders come on line and keep operating without interruptions,
- the world economy keeps strong enough during the next century – the build-up phase of the breeder systems – to support the huge investments of money (hundreds of billions of US dollars each year), materials, energy and manpower, needed for construction of some 60 breeder reactors and associated reprocessing facilities and fuel fabrication plants,
- decommissioning and dismantling of closed down facilities are ignored.

In scenario 1 a phase-out of LWR fuel reprocessing is assumed, from 2030 on. The combined gross electricity generating capacity of the first batch of breeders would be some 56 GW, 15% of the current world nuclear fleet. From 2030 on, the doubling time of the breeder system would be the pacing factor of the extension of the breeder capacity, if the LWRs were to be phased out, e.g. because of depletion of the high-quality uranium ores.
In scenario 2 the world LWR fleet is assumed to remain constant through 2070 and to be phased out after that year, because of depletion of uranium ores of sufficiently high quality. In 2100 all LWRs would be closed down. During the operating time of the LWRs all spent fuel of the world LWRs would be reprocessed from 2030 on, requiring the construction of a considerable number of large reprocessing plants. About 60 Mg plutonium each year would become available from LWRs, enough to start up 14 new breeder cycles.

None of the assumptions of the above scenarios are based on empirical facts and therefore are speculative. It should also be noted that both scenarios would require a huge new build of reprocessing and fuel fabrication facilities and of breeder reactors. Such a programme alone may pose serious logistic problems.

Proliferation and terrorism risks

MOX fuel is a mixture of uranium dioxide UO2 and plutonium dioxide PuO2. MOX fuel is at its most vulnerable during transportation and risks of sabotage and hijacking must be considered seriously. 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 or plutonium dioxide together with conventional explosives to produce a nuclear explosive are not technically demanding and do not require materials from special suppliers. The information required to carry out these operations is freely 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 the *Nth Country Experiment* proved [Frank 1967] Q591, [Schneider 2007] Q590. Nuclear weapons can be made from reactor-grade plutonium, although those made using weapon-grade plutonium are somewhat more effective. The USA and UK exploded such devices in 1956 and in the 1960s. A good nuclear weapons designer could construct a nuclear weapon from 3-4 kg of reactor-grade plutonium. Less reliability or a less predictable explosive yield than a military weapon will be no point to any terrorist group.

Safeguards

Storage and transport of MOX fuel assemblies on a scale envisaged by the nuclear industry (both Generation III and Generation IV reactors will rely on plutonium recycling) will be extremely difficult to safeguard. The risk of diversion or theft of MOX fuel pellets or fuel assemblies by personnel within the industry or by armed

and organised terrorist groups is a dreadful possbility.

The safeguards agencies claim that a commercial plutonium reprocessing plant can be safeguarded with effectiveness of about 99%. This means that, even under the most optimistic assessments, at least 1% of the plutonium throughput will be unaccounted for. Some independent experts estimate that, in practice, a more realistic figure for the effectiveness is 95% and that at least 5% of the plutonium throughput will be unaccounted for [Barnaby 2005a] Q339, [Barnaby 2005b] Q340.
What do these figures imply? A plant reprocessing 800 Mg spent fuel a year and producing about 8000 kg plutonium a year, for example the Japanese Rokkasho-Mura plant, would have a potential 'material unaccounted for' (MUF) of 80 kg (1%) – 400 (5%) kg plutonium a year.

A striking example of the problems concerning safeguards is the leak in the Brittish THORP reprocessing plant at Sellafield. A solution, containing spent fuel elements dissolved in nitric acid. leaked into a cement secondary containment chamber. The leak was not detected until April 2005, eight months after it began, by which time about 83 m^3, containing about 160 kg plutonium, leaked out. This incidence is an example of the inadequacies of the safguards system for reprocessing plants.
Measurement of the exact quantities of plutonium entering the reprocessing plant is virtually impossible, for various reasons. The operators of the reprocessing plant will be uncertain about the precise amount of plutonium produced in the plant The uncertainty is called the 'material unaccounted for' or MUF.
The same problems hold true for the thorium-uranium-233 cycle and for partitioning and transmutation, a concept aimed at the reduction of long-lived radioactive waste.

Security issues of the breeder and P&T cycles

If a breeder system were to come into operation, very large amounts of separated plutonium would be circulating in the cycle of breeder reactors, reprocessing plant and fuel fabrication plant. This would raise severe nuclear security problems. What's more the breeder cycle would generate much more high-level radioactive waste than conventional nuclear power stations and would discharge massive amounts of radioactive materials into the environment. These discharges are an unavoidable byproduct of reprocessing.

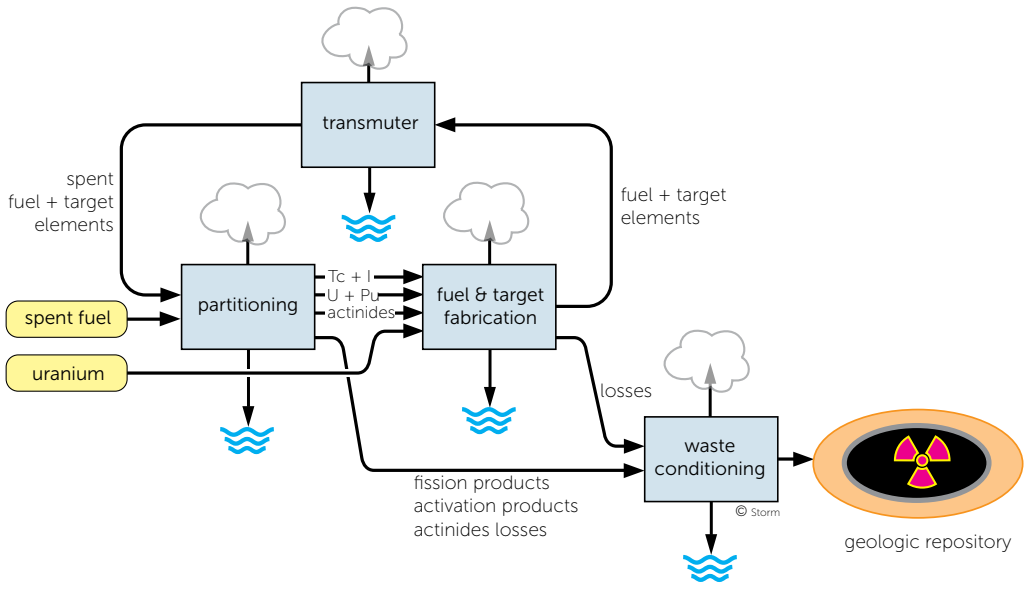

Figure 5
The partitioning & transmutation system consists of a transmutation cylce plus a waste conditioning facility and a geologic repository, Three components form the cycle: a transmuter (reactor), a partitioning (reprocessing) plant and a facility to produce fuel elements and targets containing the nuclides to be transmuted.

The radioactive wastes from the cycle are to be conditioned and stored in geologic repository. In this flowchart the inputs of chemicals, materials and energy have been omitted, as well as the waste streams arising from decommissioning and dismantling the facilities.
Operating the P&T cycle would raise the above mentioned security problems to a much greater extent than the breeder cycle, because the P&T cycle would also circulate considerable amounts of separated actinides including neptunium and americium, in addition to the separated plutonium.
Fortunately, from a safety point of view, the breeder and P&T concepts can only exist in cyberspace.

Thorium fuel cycle

The thorium breeder is based on the conversion by neutron capture of non-fissile thorium-232 into fissile uranium-233, by a similar system as the uranium-plutonium breeder. The feasibility of the thorium breeder is even more remote than that of the U-Pu breeder.
Additional roblems include:

- high radioactivity of U-233, which is always contaminated with traces of U-232,
- similar problems in recycling thorium due to the highly radioactive Th-228,
- technical problems (not yet satisfactorily solved) in reprocessing of thorium fuel.

An overview of research projects in the past and of advanced thorium reactor concepts is given in [WNA-Th 2015] Q302. Research and development on the thorium cycle has been less intensive than on the U-Pu cycle and never reached the prototype phase, like the U-Pu cycle with the French Superphénix.
India still conducts some research on thorium-U233 fuel cycle.
As [WNA-Th 2015] put it:

> 'Much development work is still required before the thorium fuel cycle can be commercialised, and the effort required seems unlikely while (or where) abundant uranium is available.'

Building up a Th-U-233 breeder system would pose a severe logistic problem. 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 more than 250 years.
Theoretically this period could be shortened to some 50-100 years, if:

1 Today's world nuclear power reactor fleet would be replaced by a new generation of LWRs which are appropiate to breed uranium-233 from thorium; the currently operating power reactors are not.
2 The spent fuel from all power reactors of the world would be reprocessed.

Both assumptions are unrealistic.

A major drawback of the thorium cycle is that a genuine thorium breeder reactor cannot sustain a fission process in itself, but always need an external accelerator-driven neutron source, or the addition of extra fissile material, such as plutonium.
Even if 370 GW of thorium breeders would come on line in 2050, their contribution to the world electricity generation capacity would be less than 7% by that time, if we assume a growth of 2% a year of the world electricity demand. The contribution to the total world energy supply would be less than 1%.

Conclusions

The breeder system has proved to be unfeasible. After six decades of intensive research it seems extremely doubtful if the theoretical and technical problems can be overcome.

Even if the breeder system would operate flawlessly from this year on, it would take about a century before the share of breeders of the world energy supply would become significant.

Any nuclear renaissance scenario for the next decades should be based on thermal-neutron reactors, mainly light-water reactors (LWRs).

The thorium-based breeder system is even more remote than the U-Pu breeder system.
Building up a thorium breeder system – provided it will ever become feasible – of an appreciable size would take centuries. Only negligible quantities of uranium-233 exists in the world.

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