

*Hello Plutonium !
Plutonium, hell!, no !*

Background Information on a Current Topic

November 2002

No Future Plutonium ?

1. Introduction
2. A few basic remarks
3. Plutonium inventories
4. What is to be done with Plutonium?
5. Economical aspects
6. How do countries involved deal with this problem?
7. Conclusion

1. Introduction

In November 1994 SPIEZ LABORATORY published a first background information addressing various plutonium-related problems¹. The main reason for doing so were increasing international smuggling activities involving radioactive materials, including plutonium. The authors dealt with questions about origin of the material, associated health risks together with questions concerning the use of plutonium in making nuclear weapons. At that time neither waste management and disposal problems nor other possible uses of plutonium were discussed. Today disposal is one of the dominant problems.

Plutonium is produced in each civil nuclear power plant in quantities of approximately one pound per day. In the course of time this yielded amounts of plutonium which in the view of many experts present a considerable proliferation risk, the risk that enough of this important material could get into the hands of would-be bomb-makers. In addition to this, the disarmament process in the nuclear weapon states, as welcome as it may be, creates large amounts of "excess"-plutonium in "weapon-grade"-quality, which also require proliferation-safe handling and storage. On the other hand, plutonium is a raw material for the production of nuclear energy.

The main questions are:

- How could it happen that so much plutonium was produced?

- Is this plutonium predominantly just waste that has to be disposed of safely, or is it a valuable raw material for further use?
- If such a further use can be found, is it profitable?
- What are the possibilities and methods for disposal?

The paper at hand tries to give some answers to these questions of current interest.

2. A few basic remarks

What is plutonium?

Plutonium is a chemical element, the element with the atomic number 94. This number describes the fact that invariably every plutonium atom contains 94 **protons** (particles carrying a positive electric charge) in its nucleus. All elements have several **isotopes**, forms of the element that are differentiated by the number of **neutrons** (electrically neutral particles with about the same mass as protons) in their nuclei. The "mass number" of an isotope refers to the sum of protons and neutrons. The nucleus of the plutonium-isotope with the mass number 239 (Pu^{239}) e.g. contains 145 neutrons in addition to the already mentioned 94 protons.

Plutonium production

In nature Pu^{239} exists only in trace quantities. All the rest is man-made, produced in nuclear reactors. Neutrons, abundant in nuclear reactors, may be "captured", if they collide with uranium nuclei. The new nuclei are energetically unstable and "decay" through an intermediate step into plutonium nuclei. Neutron capture may also happen in plutonium

¹ to be found at
http://www.vbs.admin.ch/ls/d/h_info/plutonium/
(At present available in German only.
An English version will be published in 2003).

nuclei. Fig. 1 shows the production mechanism of some plutonium isotopes. Depending on the type of nuclear reactor and the time the fuel-rods are irradiated in the reactor, the relative abundance of the different plutonium isotopes varies widely.

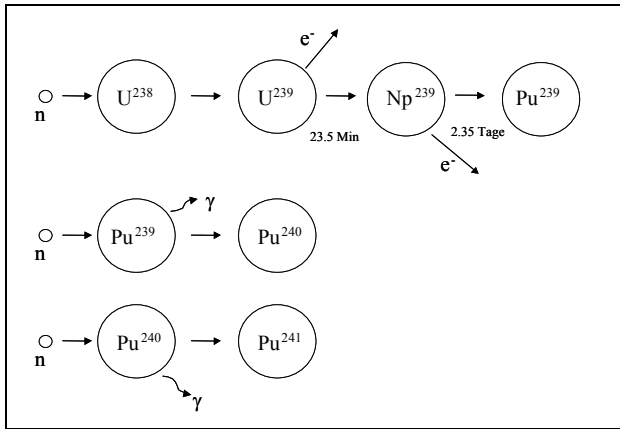


Figure 1: Neutron capture in the most abundant uranium isotope U^{238} leads to U^{239} , which decays within minutes into the intermediate nucleus Np^{239} , which in turn decays within days into Pu^{239} . Neutron capture in these plutonium nuclei leads to the higher numbered isotopes Pu^{240} , Pu^{241} and Pu^{242} . Starting from U^{235} another sequence of neutron captures and decays produces the isotope Pu^{238} .

3. Plutonium inventories

How much plutonium is there?

Around the globe the official and the "de facto" nuclear weapon states together have produced a total of about 250 metric tons of plutonium in the quality best suited for **nuclear weapons**. On 1 September 2000 the US and Russia signed a mutual agreement committing both sides to dispose of 34 tons of plutonium, most of it coming from dismantled nuclear weapons. This is more than ten thousand times the amount of plutonium that formed the core of the nuclear weapon which destroyed the city of Nagasaki. If the US and Russia actually would reduce their arsenals to a couple of thousand warheads each, this would mean that another 100 tons of plutonium would have to be disposed of.

On top of these already huge quantities of "military" plutonium have to be added the even larger amounts of **civil** plutonium. Up to now civil nuclear power plants (NPP's) have produced a worldwide total of about 1'400 tons of plutonium, a quantity which increases at a rate of about 70 tons per year. The largest part of this plutonium, about 1'200 tons, is still contained in the irradiated and thus highly radioactive so-called "spent" fuel-rods. Another approximately 225 tons are "separated" plutonium. In this case "separated" indicates, that in a highly complex mechanical and chemical process plutonium was extracted from spent fuel-rods and thus separated from all the other constituents of the spent

fuel. This process is accomplished in so-called "reprocessing plants".

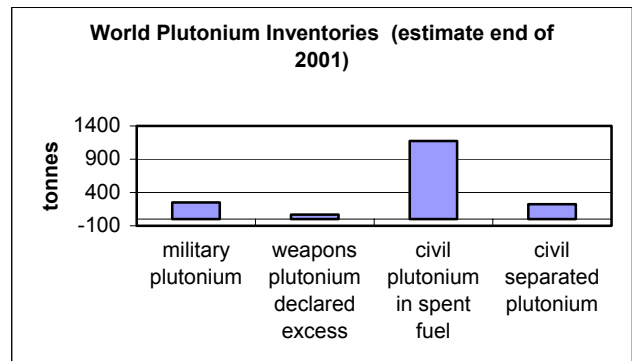


Figure 2: Global plutonium inventories at the end of the year 2001.

A side remark: These 1'400 tons would form a cube with a side length of about 4.1 meters, whereas the separated civil plutonium would fit into a cube with a side length of 2.2 meters.

Why is there so much civil separated plutonium?

In the sixties and, accentuated by the oil-crisis, in the seventies, it was generally expected that the production of electric energy in nuclear power plants would strongly increase, a process going hand in hand with a uranium shortage and accordingly also a rise in the price of uranium.

In this situation the idea came up to make the most of uranium by using so-called "fast breeder reactors". This type of reactor produces energy by fissioning plutonium nuclei, but is constructed in such a way as to generate, to "breed", new plutonium as a raw material for the future operation of the reactor by placing a uranium blanket around the reactor core. According to this concept the plutonium produced in conventional commercial nuclear power plants, mostly Light Water Reactors (LWR)², was to be separated and "burned" in fast reactors.

Another idea is to replace part of the uranium in LWR's by plutonium. Spent fuel from civil NPP's typically consists of 96% uranium, 3% fission products and about 1% plutonium. Only the fission products are just waste, the remaining 97% can be recycled to produce new fuel elements. Quite a few LWR's have started to use so-called MOX-fuel³, fuel-elements that contain a mixture of plutonium and uranium.

For these reasons, reprocessing facilities were built (Windscale 1969, La Hague 1966, Marcoule 1958, US West Valley 1966) and long-term contracts signed between NPP's and reprocessing plants to separate plutonium from spent fuel-rods.

² in Light Water Reactors ordinary water is used for cooling purposes and to slow down (moderate) neutrons. Subspecies are Boiling Water Reactors (BWR) and Pressurized Water Reactors (PWR).

³ MOX is short for **Mixed OXide**

However, the number of power plants constructed world-wide was much smaller than had been expected a few decades ago. The demand for uranium did not increase substantially either. The same holds for the military; on the contrary, large amounts of highly enriched weapon uranium are nowadays blended down to low enrichment, sold and used to manufacture fuel-rods. In addition to this the uranium deposits easily accessible for mining and milling turned out to be much greater than expected.

For all these reasons, the price of one kilogram of uranium decreased drastically from about 100 \$ in the year 1960 to about 30 \$ today (in terms of 1995's dollars).

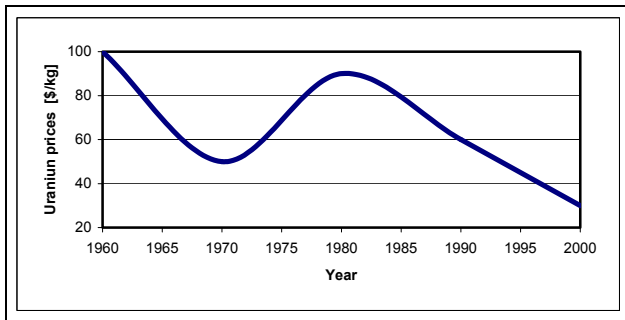


Figure 3: Development of the price of uranium in the last forty years.

Under these circumstances fast breeders are not profitable, not to mention the technical difficulties and the world-wide rejection by the public. For these reasons fast breeders have no future, neither in the US nor in Western Europe.

The experimental plants in Kalkar, Germany, in Dounreay, UK, as well as Superphénix in Creys-Malville, France were shut down. An accident that happened in 1995 put the Japanese fast breeder in Monju out of operation, maybe permanently.

Because this has been agreed upon in so-called "base-load"-contracts between nuclear power plants and reprocessing plants, the separation of plutonium from spent fuel-rods continues despite the decrease in demand. Furthermore, there were bottlenecks in the production of MOX-fuel. Not all of the nuclear power plants licensed to use MOX could get enough of this type of fuel. In the near future this will change. In October 2001 the Sellafield MOX-plant (SMP) was licensed to fabricate MOX-fuel. With this the world-wide MOX-production capacity increases by about 50%.

What are the predictions?

By renouncing from further separation and by an enhanced use of MOX-fuel a future decrease in the amount of separated civil plutonium can be expected. In June 2000 the Institute for Science and International Security (ISIS) published an estimate of the amounts of separated civil plutonium world-wide (see Figure 4).

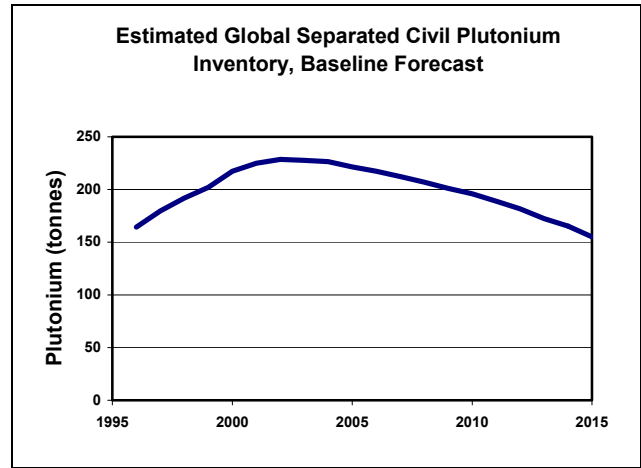


Figure 4: Estimated global separated civil plutonium inventory, baseline forecast.

There are more pessimistic forecasts, predicting on the one hand a less pronounced rise from 2002 onwards, but on the other hand seeing no decrease at all at later times.

4. What is to be done with plutonium?

There is a world-wide consent that plutonium has to be **proliferation-safe**, meaning that measures have to be taken to prevent that significant amounts of plutonium get into the wrong hands. Simplifying things a bit, it can be stated that a three-stage approach could solve the problem.

The first stage is a secure storage of plutonium for an indefinite period of time, the second stage is to "minimize its accessibility" and the third is to prevent any access for all times and possibly even to go ahead with its elimination or destruction.

Stage 1: Storage for an indefinite period of time

In the Nuclear Weapon States more than 100 tons of **military plutonium** have been declared excess to weapon requirements. Most of this plutonium is in obsolete warheads. In dismantling these warheads the plutonium-"pit" is removed and stored (see figure 5). Thus maximum flexibility is maintained with regard to future utilization or disposition. Unfortunately there is also the possibility of a rather quick re-assembly of warheads.

The credibility of disarmament efforts could be enhanced if the declared military excess plutonium were made available for safeguarding by the International Atomic Energy Agency (IAEA), in the same way as civil plutonium is controlled. Because pit-designs are highly classified, the pits probably would have to be molten down and converted into plutonium-oxide powder. At the end of the year 2000 only two tons of plutonium from nuclear weapons were placed under IAEA-safeguards which means that the IAEA knows the whereabouts of his material. The Agency issues prescriptions about storage and physical protection and periodically carries out inspections.

In separated **civil plutonium** from power reactors long-term storage creates additional problems.

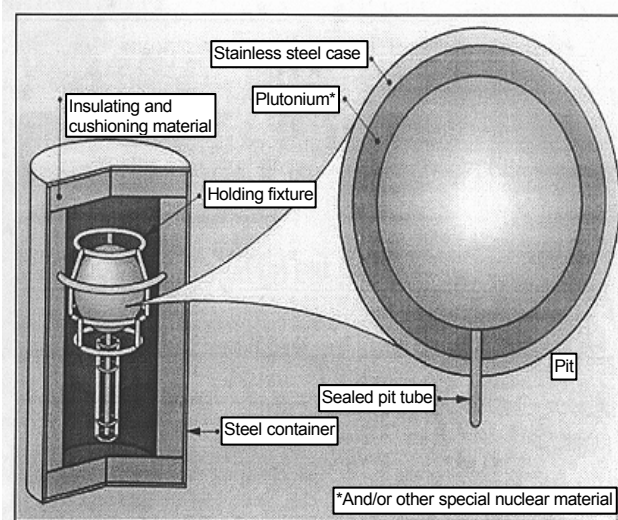


Figure 5: Drawing of a storage container for plutonium-pits. In the Pantex plant in Amarillo, Texas, which is part of the US nuclear weapons complex, more than 12'000 such pits are kept under rigorous security and safety measures.

Reactor-grade plutonium contains much more Pu^{241} than weapon-grade plutonium. This Pu^{241} decays rather quickly into americium-241 (Am^{241}), a strong emitter of gamma-radiation. Already after a few years of storage this radiation makes handling difficult and a direct use of reactor-plutonium for the production of fuel rods impossible. In order to fulfil the specifications of MOX-production facilities, americium and plutonium would have to be separated chemically (costs 1994: between 10 \$ and 28 \$ per gram of plutonium). Separated civil plutonium poses a proliferation risk, too and therefore has to be extremely well safeguarded and physically protected. The costs of a secure storage are estimated to be between 1 \$ and 5 \$ per gram of plutonium and per year.

Stage 2: "minimized accessibility"

In 1994, the US National Academy of Sciences defined the minimum requirements to make weapons plutonium inaccessible, the so-called **spent-fuel standard: "Make plutonium roughly as inaccessible for weapons use as the much larger stock of plutonium in civilian spent fuel"**.

It is true that plutonium is radioactive, but the emitted radiation is mostly alpha-radiation with a strongly limited reach. Beta- and gamma-radiation are also emitted, as well as neutrons, but even the cumulated radiation is too weak to "self-protect" plutonium from being diverted or stolen. In contrast to this, the gamma-radiation of the fission products in spent fuel-rods is, even after decades, still a hundred to a thousand times stronger. Thanks to this radiation, plutonium in spent fuel is much better protected than separated plutonium, because handling of this type

of material requires extended shielding and radiological protection measures.

The definition of the spent fuel standard is somewhat hazy. After all there are different types of reactors, different compositions of the fuel, as well as different burn-ups⁴. As a consequence also the radiation properties of the spent fuel-rods vary widely. By consensus the "standard" spent fuel is assumed to be 30 year-old fuel from a LWR with a burn-up of 33'000 [MWd/t]⁵.

Basically there are two ways to achieve this spent fuel standard, one is to "burn" **MOX** in a reactor, and the other one is **immobilization**.

A: MOX (Mixed Oxide)

MOX-fuel rods comprise mostly uranium-oxide and only 6% to 7% plutonium-oxide. In modern LWR's about one third of the uranium fuel elements can be replaced by MOX-elements. In the MOX-rods more plutonium is fissioned than newly created by neutron capture in the uranium part of the rod. This decrease is more or less compensated for by the production of plutonium in the normal uranium-oxide-rods. The point is, that after burn-up the remaining plutonium in the MOX-elements and the newly produced plutonium in the uranium-oxide-elements are now immersed in highly radioactive fission products and thus correspond to the spent-fuel standard. Plutonium in irradiated MOX-elements has an isotopic composition very unfavourable for both weapon purposes and another cycle in a nuclear reactor.

The MOX-solution is not without controversy. Some arguments for, and against the use of MOX, respectively, see next page.

B: Immobilization

"Can in canister" is one of several immobilization methods. Thereby plutonium is embedded in chemically stable ceramic material in the shape of flat cylinders about one inch high and three inches in diameter. These disks are stacked in a canister which afterwards is filled with a mixture of molten glass and high level waste (highly radioactive fission products). This vitrification, together with a subsequent underground long-term storage of the canisters, establishes the desired immobilization. Summing up: Once separated at great costs, plutonium is now reunited with fission products, again at great costs.

Which one of the two ways to achieve the spent-fuel standard, MOX or immobilization, will turn out to be less expensive, faster and more reliable and therefore better suited to make plutonium proliferation-proof, cannot be assessed once and for all. Most countries have decided or probably will decide in favor of the MOX-process.

⁴ **burn-up** indicates how much energy was produced with a given amount of fuel. The higher the burn-up, the more fission products are contained in the spent fuel.

⁵ A common unit to describe burn-up is Megawatt days per ton of fuel [MWd/t].

Arguments speaking for MOX
<ul style="list-style-type: none"> - Plutonium stocks are a huge energy potential that ought not be thrown away. - The proliferation-risk can be diminished by "burning" MOX containing weapon-grade plutonium. - There are more than 30 years of experience with MOX without any serious accidents.
Arguments speaking against MOX
<ul style="list-style-type: none"> - There is a need for more transports of proliferation-prone plutonium. - The total amount of plutonium in the reactor is greater, therefore the consequences of a grave accident would be worse. - Because plutonium is more radioactive than uranium and because plutonium has to be safeguarded, the production of MOX-fuel is more expensive than the production of uranium-fuel. - Because of its composition spent MOX-fuel is more radioactive and produces more heat for a longer period of time. Therefore, spent MOX has to be kept in intermediate storage for about 150 years as compared with only about 50 years for spent uranium fuel.

If the process should stop at this stage and independently whether immobilized or used as MOX in a reactor, the problem eventually comes up to find suitable repositories for a safe final disposal of "spent fuel standard"-plutonium. Up to now in all the countries concerned only intermediate or long-term storage facilities are in existence.

Yucca Mountain in Nevada could become the very first geological final repository for civil radioactive waste. It is scheduled to start operation in the year 2010, but today this seems hardly possible, because of the widespread opposition not only in Nevada but throughout the US.

Stage 3: Prevent any access for all times

"Spent-fuel standard"-plutonium brings about a smaller proliferation risk than the separated variety. But all the plutonium is still there and could be separated (once more in some cases) from its associated highly radioactive fission products.

If the goal were to surpass the "spent fuel standard", then this should hold not only for the separated civil and excess military stocks, but, to be consistent, also for the much larger amounts contained in the spent fuel-rods. Can this be realized? Can plutonium be protected infinitely not only against unauthorized access but against any access at all?

Ideas to achieve this are manifold, including crazy and even ludicrous propositions. First, a list of

methods that do not destroy the plutonium but rather put it out of reach:

- shoot it into outer space.
- shoot it into the sun.
- distribute it over the seven seas.
- arrange it deep underground around a nuclear weapon. The detonation of this atomic bomb distributes the plutonium in the surrounding rocks.

For a lot of reasons none of these ideas are seriously taken into consideration. Pursuing them would be too dangerous for man and environment, too expensive, too slow...

Fission is the only way to destroy plutonium and this can only be achieved in a reactor, in reactor-like systems or by means of a particle accelerator.

Below some of the possible methods:

- Light Water Reactors

In order to destroy plutonium in LWR's, more than one third of the fuel elements have to be MOX-elements. It is possible to construct LWR's where 100% of the rods are MOX-rods. To control the neutron flux in these reactors additional and more efficient control rods are required than in conventional LWR's. The slow neutrons, predominant in this reactor type, split only the odd-numbered plutonium isotopes (Pu^{239} , Pu^{241} , Pu^{243}). The even-numbered isotopes (Pu^{238} , Pu^{240} , Pu^{242}) are fissionable by fast neutrons only. In order to destroy them in a LWR, they first would have to be transmuted by neutron irradiation into odd-numbered isotopes.

- Fast reactors (liquid metal reactors, fast breeders)

In these reactors operating modes are possible where more plutonium is fissioned than newly produced. Thereby, the predominantly fast neutrons – responsible for the word "fast" in this reactor type – also split the even-numbered isotopes. This would be an advantage, but for reasons described above it is unlikely that fast reactors will play a role in the process of plutonium destruction.

- Non fertile fuel or inert matrix fuel (IMF)

In these systems uranium in the fuel rods is replaced by other materials, thus preventing the production of new plutonium. Apart from the thermal properties, also the neutronic properties of the substituting material should closely match the ones of MOX- and/or uranium-oxide-elements. Very promising experiments were performed in LWR's with single fuel elements made up from oxides of zirconium, plutonium, yttrium and erbium, but there is no power reactor yet, which uses exclusively uranium-free fuel elements.

- Other methods

like for example accelerator-driven systems or high-temperature gas-cooled reactors are relatively new concepts, whose development, realization and licensing would take up decades.

In all the methods described above, a virtually complete destruction of plutonium would time and again call for reprocessing. This repeated reprocessing, transporting and recycling of plutonium would increase the proliferation risks and the risks for man and environment for decades, maybe even for centuries, before reducing them below today's level.

5. Economical aspects

The costs of a complete or almost complete destruction of plutonium strongly depend on the method chosen. In this context estimates are extremely difficult. However, there is reliable and comprehensive information about the costs of destroying some plutonium by a single cycle of reprocessing fuel-rods, recycling plutonium in the form of MOX and "burning" it in a LWR.

In June 2000 a study was published that had been ordered by the former French prime minister

Lionel Jospin. This study investigated material flows, waste problems and financial expenditures of the French nuclear-energy complex. In France 65 to 75% of the spent fuel are reprocessed and the major part of the separated plutonium is recycled in the form of MOX and "burned" in 20 out of the 58 French LWR's. The study shows that by doing so the amount of plutonium that has to be disposed of can be reduced by about 15 to 17%.

France could produce its electric energy much cheaper without this cycle of reprocessing and subsequent use of MOX. A comparison of all the costs incurred in the production of nuclear energy, starting with the production of fuel elements up to waste disposal, shows astonishing and impressive results: Every kilogram of plutonium that, thanks to reprocessing and recycling, is destroyed and thus does **not** have to be disposed of otherwise, costs the French consumer and tax-payer about 200'000 Euro.

6. How do countries involved deal with this problem?

USA	Based on a decision taken by President J. Carter in 1977, still in force, the US do not reprocess civil spent fuel-elements. Disposition of the 34 tons of military excess plutonium agreed upon with Russia, was originally scheduled to take place in two ways: 8.43 tons were to be immobilized and 25.57 tons to be "burned" in reactors. The US wanted to keep open both paths, on the one hand because this expedites the process, and on the other hand because this allows to remain flexible if, for whatever reasons, one of the two methods should run into difficulties. The Bush Administration now favors the MOX-path. In March 2001 further development of immobilization techniques was suspended for the time being and in January 2002 it was decided to transform the whole 34 tons into MOX.
Russia	Russia considers plutonium a valuable source of energy and dislikes to "throw away" its 34 tons of military excess plutonium. Because disposition of this military plutonium has been given priority, Russia will not be able to diminish its stock of civil separated plutonium (about 30 tons).
UK	The Sellafield plant alone stores about 60 tons of plutonium. Since fast breeders are no longer an option, this large amount of plutonium is the reason for growing concern among authorities, politicians and the population. About one quarter of the total electric power today comes from NPP's. This part will decrease to about 3% in the year 2020 and no more plutonium will be "burned". Since the MOX-path would require new modern NPP's, the immobilization path has a good chance.
France	In France 59 NPP's produce about 75% of the total electric power. About two third of the spent fuel are reprocessed and the largest part of the separated plutonium is recycled in MOX-form in about 20 NPP's. The exorbitant costs make sure that withdrawal from reprocessing is at least taken into consideration.

Japan	Japan is very much interested in the MOX-solution and plans to load all his LWR's with MOX-fuel. Until the year 2010 up to 20 NPP's should be able to "burn" MOX.
Germany	Since the seventies, German NPP's have collected experience on MOX-fuel. At the time being, 11 of the 12 licensed NPP's "burn" MOX. An attempt of the German government to withdraw from current reprocessing contracts failed because of the opposition from France and the UK. After July 2005 however, fuel-rods will no longer be transported and reprocessed abroad. On 14 December 2001, the German parliament, the "Bundestag" consented to the "Gesetz über den Atomausstieg", the law regulating the withdrawal from nuclear energy. Thus, step by step the 19 German NPP's will be shut down in the course of the next 20 years.
Switzerland	<p>Switzerland uses MOX in three (Beznau I, Beznau II and Gösgen) out of its five NPP's. At the end of the year 2000 the Swiss plutonium inventory was as follows:</p> <ul style="list-style-type: none"> - 0,6 tons in non-irradiated MOX. - 8 tons in spent fuel in intermediate storage at the reactor sites. - 3 tons in spent fuel sent abroad for reprocessing. <p>A first draft of the (future?) nuclear energy-law forbade reprocessing. The first of the two chambers in the Swiss government, the "Ständerat", reduced the ban to a ten-years moratorium. In June 2002 the second chamber, the "Nationalrat", declared itself in favor of reprocessing, albeit only under stringent restrictions. After both chambers will have agreed upon the final version, the Swiss population will vote on the bill. The referendum will probably take place at the end of the year 2003.</p>

7. Conclusion

From a financial point of view, reprocessing and recycling of spent fuel are not worthwhile at all. The reduction of the amount of plutonium that has to be disposed of as waste is disappointing. Uranium is abundant and cheap. Since fast breeders are certainly not an option for the next decades, there is no incentive to separate plutonium. On the one hand, it is true that the risk of theft and subsequent misuse of separated plutonium is almost negligibly small, but on the other hand, this risk is not zero and

it increases with increasing amounts of separated plutonium as well as with an increasing number of transports and shipments of plutonium between reprocessing plants, storage sites, MOX-fuel production sites and reactors.

With the existent and planned facilities it would take up decades until all plutonium would correspond to the spent fuel standard, even if all reprocessing were stopped right away.

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