

Revisiting the thorium-uranium nuclear fuel cycle [DOI: 10.1051/EPN:2007007]

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THE thorium-uranium nuclear fuel cycle, in which the main fissile nucleus is uranium 233 and fuel regeneration is ensured through neutron capture on thorium 232 offers many potential advantages as compared to the better known uranium-plutonium fuel cycle. These include, in particular, reduced high activity long lived waste production and less likelihood of nuclear proliferation. A brief description of the nuclear reactors being considered for this fuel cycle is given. We show also that a strategy can be put together to constitute the initial uranium 233 supply for such reactors, using today's pressurized water reactors and a thorium and plutonium mixed oxide fuel.

Introduction

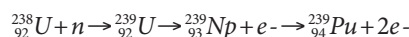
Today's reactors burn essentially uranium 235; they use only about 1% of the natural uranium [1]. For this reason, uranium reserves are estimated to provide about a century of reactor operation, the actual time span depending on the number of reactors in operation in the world and on the accepted cost of natural uranium. In the 1950's, because of these relatively small reserves, reactor physicists proposed to develop *breeder* reactors, in which the main fissile nucleus is no longer uranium 235 but either plutonium 239 or uranium 233 instead. Indeed, when these nuclei fission, they emit enough neutrons to ensure their replacement (breeding) through neutron capture on uranium 238 or thorium 232 respectively. Neither plutonium 239 nor uranium 233 can be found on earth in significant amounts so that they have to be produced if reactors using them are to be developed. Plutonium 239 is produced automatically in almost all of the reactors that are being operated worldwide, since these burn uranium-based fuels. The availability of large amounts of plutonium led to the development of the so-called uranium-plutonium fuel cycle, a concept that was realised in France with the Phenix and SuperPhenix breeder reactors. By contrast, the

amounts of uranium 233 available remained tiny, insufficient to allow the rapid development of a thorium-uranium concept.

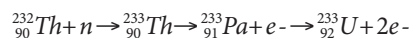
It appears today that the growth rate of nuclear power worldwide does not require the fast development of breeder reactor concepts. It is then possible, as we show in this paper, to constitute a stockpile of uranium 233 that could allow the development of a fleet of thorium-uranium reactors. We show also that such a concept has many major advantages, in particular concerning the disposal of radioactive waste and the risks of nuclear proliferation. We give a brief description of the types of reactors being considered to implement this fuel cycle and of a strategy that makes use of today's reactors to create the initial stockpiles of uranium 233.

The advantages of the thorium fuel cycle

Breeder nuclear reactors such as SuperPhenix are based on the uranium 238-plutonium 239 fuel cycle. In this cycle, the plutonium, whose fission is the source of energy released in the reactor, is replaced by the new plutonium obtained through the capture of a neutron by a uranium 238 nucleus:



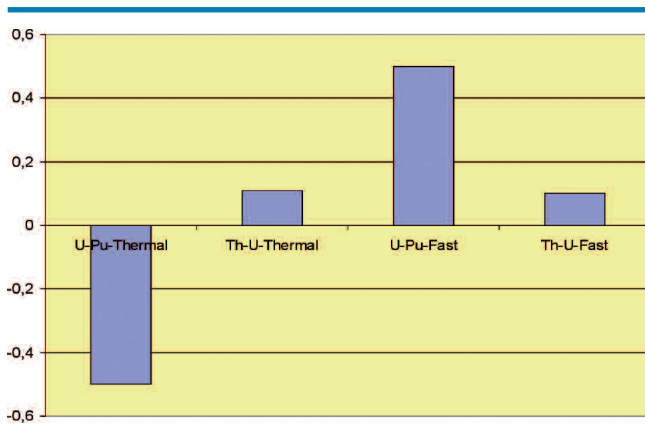
In the thorium 232-uranium 233 cycle, thorium 232 plays the role of uranium 238 and uranium 233 that of plutonium 239:



As Figure 1 illustrates, while the uranium 238-plutonium 239 fuel cycle requires fast neutrons to be sustainable, the thorium 232-uranium 233 fuel cycle is sustainable with either thermal neutrons or fast neutrons.

A small initial inventory

The probability that the fission of a fissile nucleus (²³⁹Pu or ²³³U) will occur relative to the probability of a simple neutron capture on a fertile nucleus (²³⁸U or ²³²Th) is larger with slow neutrons than with fast neutrons. As a result the amount of fissile nuclei necessary for a reactor with slow neutrons to operate is usually smaller than that needed for a reactor with fast neutrons. Thus, in the case of the uranium 238-plutonium 239 cycle, the amount of ²³⁹Pu required to operate a fast neutron breeder reactor with a thermal power output of 3 GW is typically on the order of 7 to 14 metric tons [2] (depending on the amount of plutonium held up in the reprocessing system, the in-core inventory being on the order of 7 metric tons). In the case of the thorium 232-uranium 233 thermal cycle, the amount of ²³³U required to operate a slow neutron breeder reactor with the same power output is only 1.5 to 3 metric tons [3]. As a result, the production (by thorium 232 irradiation in a "classical" reactor) of the initial uranium 233 load for a thorium-uranium breeder reactor would be four times shorter than the production (by uranium 238 irradiation in the same type of reactor) of the initial plutonium 239 load for a uranium-plutonium breeder reactor.



▲ Fig. 1: Number <n> of neutrons available for breeding in the uranium-plutonium and the thorium-uranium cycles with thermal and fast neutron spectra. Breeding is impossible for negative values of <n>.

Reduced Radioactive Wastes

After a few centuries, the radiotoxicity of radioactive wastes is due mainly to that of the heavy ($Z > 92$) alpha emitting radioactive nuclei that are produced by successive neutron captures in the heavy elements present in the nuclear fuel. In breeder reactors based on the uranium-plutonium cycle, the uranium and plutonium are normally completely used up after fuel processing, they contribute very little (leakage during fuel processing) to the radiotoxicity of the wastes which is, because of this, two orders of magnitude smaller than in the case of classical PWR [4] reactors as illustrated in Figure 2.

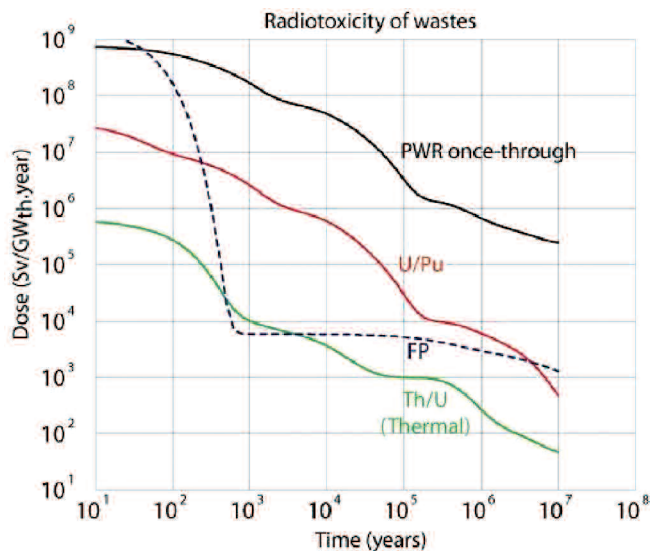
In other words, for an equal amount of energy produced, the geological storage needs will be nearly a hundredfold smaller with a uranium-plutonium based breeder reactor fleet than with the present day scheme. The thorium-uranium fuel cycle is even better than this perspective because, since the mass number of thorium 232 is 6 units less than that of uranium 238, the production of minor actinides (neptunium, americium, curium) which are the major contributors to the radiotoxicity of the wastes in the uranium-plutonium cycle is drastically reduced. Figure 2 shows that the radiotoxicity of the wastes in the thorium-uranium cycle is much smaller than that of the uranium-plutonium cycle during the first 10 000 years. It is noteworthy that the reduced initial radioactivity in this cycle would allow large savings on the size and, as a consequence, the cost, of geological storage [5].

A Non-proliferating Fuel

An important condition for the manufacture of a nuclear weapon is that the fissile elements have low intensity gamma decay since the presence of high gamma activity would require very thick lead or lead-glass protections behind which operators would have to work. The uranium 235 obtained from isotopic separation plants has a low intensity gamma emission, just as the plutonium retrieved from reactor spent fuel. In general, the production of uranium 233 entails also the production of uranium 232 whose half life is 70 years. Successive alpha decays of uranium 232 lead to thallium 208 which decays with the emission of a very deeply penetrating 2.6 MeV gamma ray. It is thus practically impossible to manufacture a uranium 233 based weapon in the presence of uranium 232 contamination. True, this advantage regarding proliferation has its counterbalance in the more complex reactor fuel handling and manufacture. The entire process has to be automated or has to be executed behind heavy shielding.

Large reserves

The thorium terrestrial reserves are estimated to be about four times those of uranium. Specifically, India, Brazil, Madagascar boast of large and very rich thorium beds. By contrast, thorium is not very soluble in water so that its extraction from sea water is not being considered, contrary to that of uranium. We should

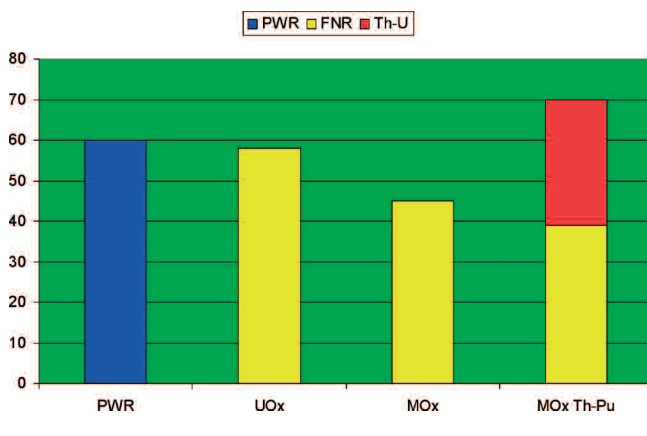


▲ Fig. 2: Time evolution of the radiotoxicity of high activity radioactive wastes per unit energy produced by the reactor (Thermal GigaWatts x year) for various fuel cycles. Nuclear wastes are due on the one hand to the actinides leaked during fuel processing (their amount depending on the type of reactor concerned) and on the other to the fission products (their amount depending only on the amount of energy produced). "At equilibrium" applies to closed cycle reactors (U/Pu and Th/U breeders; it means that the condition has been reached where equal amounts of minor actinides are created and destroyed in the reactor.

stress, however, that breeder reactor technology being very thrifty in its use of the fuel, even low content ore could be worked profitably, ensuring that fuel would remain available over several thousand years both for uranium and thorium based breeder reactors. The fact that thorium reserves are larger is thus not an important issue.

What reactor types for the thorium fuel cycle?

With the thorium-uranium fuel cycle, breeding can be achieved with fast neutrons and also with slow neutrons. With fast neutrons, it is more tricky than with the uranium-plutonium fuel cycle: the initial uranium 233 inventory is large, on the order of 5 metric tons for a 3 GW thermal power reactor. This represents the in-core inventory, to which the uranium in the fuel reprocessing system should be added (uranium in the processing unit and uranium on standby pending reprocessing). It follows that the doubling time [6] becomes quite large. As a consequence, fast neutron thorium-uranium reactors will be iso-breeders in practice. They could be similar to the fast neutron reactors based on the uranium-plutonium fuel cycle, for example those with a molten metal (sodium or lead) coolant.



▲ **Fig. 3:** Number of breeder reactors that could be started after a fleet of 60 GWe PWRs have operated over 40 years. In blue, the number of PWRs. In yellow, the number of uranium-plutonium breeder reactors that could be started with the plutonium from a UOX fuel fleet without MOX, from a classical MOXed fuel fleet burning uranium-plutonium MOX, and from a thorium-uranium MOXed fleet. In red, the number of thorium-uranium breeder reactors that could be started from the uranium 233 produced in the latter case.

Thorium-uranium breeder reactors with slow neutrons need only a small uranium 233 inventory, on the order of 1 metric ton. Their theoretical doubling time is similar to that of uranium-plutonium fast neutron breeder reactors. However, fission products are much more efficient in poisoning slow neutron reactors than fast neutron reactors. Thus, to maintain a low doubling time, neutron capture in the fission products and other elements of the structure and coolant have to be minimized. An elegant theoretical solution to this problem was proposed in the 1960's, namely, a reactor in which the fuel is a molten salt which also serves as the coolant. Neutron capture on the fission products would be limited thanks to on-line salt recycling, at the cost of additional complexity since the reactor becomes also a chemistry factory. Giving up the low doubling time objective opens the way to molten salt reactors with drastically simplified on-line fuel processing or to other reactor types, such as those with in-operation fuel loading/unloading such as heavy water reactors (CANDU) or gas-cooled pebble-bed reactors. A particularly interesting scheme would consist in complementing a thorium-uranium reactor fleet with fast neutron reactors with a uranium-plutonium core surrounded with a thorium blanket that could produce the uranium 233 needed in excess to extend an existing thorium-based reactor fleet.

Advantages and drawbacks of uranium-plutonium MOX fuels

Following the 1973 oil crisis, French experts had anticipated a rapid development of worldwide nuclear power production similar to the policy that France was about to implement. There would then be a risk that uranium resources, which are poorly utilized in PWR type reactors, would be insufficient. As a consequence, France, taking advantage of the experience acquired on the Rapsodie experimental reactor and the Phenix prototype reactor (first criticality 1967 and 1973 respectively), set out resolutely to develop sodium-cooled fast neutron breeder reactors (FNR) with the construction of Super-Phenix (initial decision in 1973, beginning of construction in 1976, first

criticality in 1985). In addition, the creation of a plutonium separation unit (La Hague) and of a plutonium loaded oxide fuel manufacturing unit was decided, these being necessary for the supply and manufacture of the breeder reactor cores.

The anticipation of the French experts did not come true, on the one hand because of the development of coal burning power plants, whose investment costs are less than those of nuclear power plants and on the other because of the Three Mile Island accident which put an end to the construction of nuclear power plants in the United States. Thus, the economic argument in favor of FNRs vanished. At the same time, their development was confronted with the virulent opposition of the anti-nuclear movement which saw them, and rightly so, as fulfilling the condition for sustainable nuclear power. The political decision that led to the shutdown of Super-Phenix, supported by the technical difficulties encountered during the first years of the reactor's operation [7], put an end to the FNR program in France at least in the short and medium term. In order to make the best of the considerable technological and financial investments as well as the human resources placed in the La Hague and Marcoule factories, the decision was made to burn the extracted plutonium in the PWR reactors. This is the MOX program in which the fuel is a mixed uranium and plutonium oxide whose composition is near stoichiometry: $(U_{1-x}Pu_x)O_2$ with $x=0.05$.

There are undisputable advantages to the MOX program: less uranium 235 consumed, considerably smaller volume of the high-activity long-lived wastes and their conditioning in very stable glass matrices, maintenance of a technical and industrial know-how which is unique in the world and constitute an exceptional asset for France in the advent of renewed nuclear power development in the world.

But there are also drawbacks to the MOX program. Irradiated MOX fuels are set aside with the idea that the plutonium they still contain could be used in future reactors. But spent UOX [8] fuels contain more plutonium and of better quality before their reprocessing than the irradiated MOX fuel thus set aside. Burning MOX fuel, then, jeopardizes the ability to constitute the fuel stockpiles needed to deploy a fleet of FNRs in the future. In addition, irradiated MOX fuels are about 5 times more radioactive than irradiated UOX fuels, making their surface storage and, even more so, their permanent geological storage more difficult.

Advantages of thorium-plutonium MOX

Today's MOX fuels are comprised of approximately 5% plutonium for 95% natural or depleted uranium. While the irradiation tends to decrease the initial amount of plutonium, the presence of uranium 238 leads to a partial reconstitution of the plutonium stockpile. The destruction of plutonium can be speeded up by replacing the uranium in the MOX with thorium, the result being thorium-plutonium MOX.

Today, a PWR burning uranium-plutonium MOX incinerates approximately 544 kg plutonium per year (for the simplicity of the argument, we assume here a "fully MOXed" PWR while, in fact, only one third of the fuel of a PWR is "MOXed"). A "fully MOXed" PWR loaded with thorium-uranium MOX should produce approximately 280 kg of uranium 233 per year while it incinerates approximately 800 kg of plutonium, producing about 20% less minor actinides. However, the quality of

the plutonium unloaded, as measured by the relative amount of fissionable plutonium isotopes, would be somewhat degraded. These results are summarized in Table 1.

Nucleus	At loading (Kg/8TWhe)	MOX Production (Kg/8TWhe)	Th Mox Production
²³³ U	0	0	278.72
²³⁹ Pu	1008	-500	-650
Total Pu	1900	-544	-800
M.A.	0	139	119

▲ **Table 1:** Comparison of the effect of MOX and thorium MOX fuels. The plutonium loaded comes from reprocessed UOX fuel and the small proportion of uranium 235 remaining in the uranium-thorium MOX is neglected. (Results from an "order of magnitude" simulation of a 1 GWe reactor operating 91% of the time).

The inventory of a fast neutron uranium-plutonium 1 GWe (electric GigaWatt) breeder reactor is approximately 7 to 14 metric tons. That of a slow neutron thorium-uranium 233 1 GWe breeder reactor would be approximately 1.5 metric tons. The plutonium incineration using uranium-plutonium MOX in a PWR would thus destroy 8% of the initial inventory of an FNR [9], while plutonium incineration with thorium-plutonium MOX in a PWR produces approximately 18% of the initial inventory of a thorium-uranium 233 [10] breeder reactor. The development of a breeder fleet, then, is not jeopardized if plutonium incineration is done with thorium-plutonium MOX. Moreover, since plutonium is no longer produced from the uranium, the net amount of plutonium incinerated would be increased by 150 kg. In Figure 3, these considerations are illustrated in the case of a PWR reactor fleet partly loaded with thorium MOX fuel (assuming that all the plutonium produced in UOX fuels is recycled in thorium MOX). In this hypothesis, the number of thorium-uranium breeder reactors obtained is significantly larger than the number of uranium-plutonium reactors "lost" because of plutonium incineration. As mentioned in the introduction, one of the reasons which disqualified thorium-uranium breeders as compared to uranium-plutonium breeders was, precisely, the non-existence of a uranium 233 stockpile to start with, while the PWR, BWR heavy water, and graphite moderated reactors were producing large amounts of plutonium. The advent of thorium-uranium MOX fuels would change the picture completely. Moreover, if, in the end, there were to be no breeder reactor program, the uranium 233 could be burned in standard reactors without entailing the production of large amounts of plutonium or minor actinides.

Conclusion

It would be unfortunate if orientations were to be decided on the basis of fast decisions made without true scientific and technological debates concerning future reactors and fuel cycles, using the argument that different orientations would break away too radically from currently accepted procedures. There is no real hurry for the definition of the systems of the future and the times lend themselves to open and thorough reflection. Granting, however, that pencil and paper designs are not sufficient, small scale prototypes of the more promising reactors will have to be built within the frame of international collaborations.

It would thus be worthwhile to resume the molten salt reactor program that was started in the 1960s at the Oak Ridge National Laboratory in the US, with the construction of a reactor prototype with a power capacity of a few ten MW. Likewise, there seems to be no valid reason to prevent the in-reactor irradiation of a few thorium MOX fuel assemblies. Finally, France should try to initiate a collaboration with Canada and India with a view to evaluating the potential of CANDU reactors vis-à-vis the thorium-uranium 233 fuel cycle. ■

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For further informations see:

- 1) H.Nifenecker, O.Méplan and S.David, *Accelerator Driven Subcritical Reactors*, IOP (2003) and references therein
- 2) Luc Mathieu, *et al.*, "The thorium molten salt reactor : Moving on from the MSBR", *Progress in Nuclear Energy* **48** (2006) 664-679
- 3) <http://lpsc.in2p3.fr/gpr/gpr/index.htm>

Notes

- [1] Natural uranium contains 0.7% uranium 235 and 99.3% uranium 238. The fuel of French PWR reactors is uranium, slightly enriched to 3.5% in uranium 235. 60% of the fissions in the reactor are due to uranium 235 and the rest are due essentially, and indirectly, to uranium 238 via plutonium 239 resulting from neutron capture on uranium 238.
- [2] In a uranium-plutonium breeder reactor, the initial plutonium load amounts to 15 % to 18 % of the initial uranium load.
- [3] In a thermal thorium-uranium breeder reactor, the initial uranium 233 load amounts to 1.5 % of the initial thorium load.
- [4] PWR: Pressurized Water Reactors, the only reactors operated commercially in France and the most widespread worldwide. BWR: Boiling Water Reactors, the second most widespread reactors having similar fuel use properties.
- [5] Without taking into account the need to handle the stockpiles that exist today and those that will be produced by the next generation of European Pressurized Reactors (EPR).
- [6] The doubling time is the time needed to produce as much uranium (or plutonium respectively) as the initial amount needed, i.e. obtain the equivalent of two initial loads starting from one initial load.
- [7] See (in French) « Super-Phénix, pourquoi ? » by G. Vendryes, Ed. Nucléon (1997) and, on the site of the French Physics Society (SFP): http://sfp.in2p3.fr/Debat/debat_energie/Nucleaire/Reacteurs/superphenix.html
- [8] The initial composition of UOX fuels is uranium oxide, UO₂ (with a c.f.c. structure of the CaF₂ type) where the uranium is slightly enriched. After irradiation, they contain the fission products, plutonium and minor actinides (neptunium, americium, curium). Reprocessing consist in extracting the plutonium and uranium and mixing the other elements in a glass matrix. As irradiation levels increase, the proportion of fissile isotopes in the plutonium decreases (especially plutonium 239) with a corresponding increase in the non-fissile isotopes of plutonium (especially plutonium 240). This constitutes an advantage vis a vis proliferation but a drawback for a reactor fuel.
- [9] Really, 544/7000 where 544 is obtained from Table 1.
- [10] Really, 278/1500 where 278 is obtained from Table 1.