Impact of the MSBR concept technology on long-lived radio-toxicity and proliferation resistance

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Abstract. The MSBR (Molten Salt Breeder Reactor) was an industrial project designed at the beginning of the seventies at Oak Ridge National Laboratory and based on Thorium. Just before, the MSRE worked very well during four years with molten fuel. The MSBR system, where a maximum breeding was wanted, included a graphite moderated core with the circulation of a 71.7%LiF-16%BeF_2-12%ThF_4-0.3%UF_4 salt and a pyro-chemical reprocessing unit. To obtain a maximum breeding ratio, Protactinium was extracted and stored allowing decay out of the neutron flux. This required the entire salt volume to be reprocessed in ten days, the gaseous fission products and Minor Actinides being extracted continuously by helium bubbling and pyro-chemical methods. The doubling time was evaluated to around 25 years. The project has since been re-evaluated especially within the frame of the EURATOM concerted Action MOST. To have an acceptable global reactivity feedback coefficient, studies have shown various possibilities based on core geometry, neutron moderation ratio and salt composition. When requiring only a breeding ratio of one, it is possible to avoid continuous reprocessing and to strongly simplify it. These various options will be discussed. The detailed inventory will be given showing clearly the interest of the Thorium Molten Salt Reactor where the production of Americium and Curium is a factor of one hundred lower that for the U-Pu RNR. The amount of Uranium 232 which is always produced in the Thorium cycle will be calculated as well as its decay rate since its decay chain eventually results in a 2.6 MeV γ –ray which may be used to detect and hence control the U233 fuel movements. As the U233 has to be produced in other reactors (PWR, RNR or other MSR), special cares have to be taken and will be discussed.

1. Introduction

Until now, nuclear energy production is based on Uranium 235, the only fissile nucleus still existing in nature. It represents only 0.7% of the natural Uranium and resource problems are unavoidable if world nuclear energy production seriously increases. The solution to massively extend the capabilities of nuclear fission is to use the only two fertile nuclei existing on the earth in reactors designed to be able to breed at least as much fissile material as they burn. After one neutron capture and two β decays, the fertile nuclei, Uranium238 and Thorium 232 are transformed into fissile nuclei as follows:

$$n + {}^{238}U \longrightarrow {}^{239}U \xrightarrow{\beta} {}^{239}Np \xrightarrow{\beta} {}^{239}Pu$$

$$n + {}^{232}Th \longrightarrow {}^{233}Th \xrightarrow{\beta} {}^{233}Pa \xrightarrow{\beta} {}^{233}U$$

The information is summarised in the actinide chart shown in figure 1. The Uranium cycle is already used in present reactors where Uranium only slightly enriched in ^{235}U and mainly composed of ^{238}U leads to the production of Plutonium. That Plutonium is partly burnt during the reactor operation, for example in the PWR's, one third of the energy is coming from the Plutonium fission. The unavoidable

production of Plutonium and other Minor Actinides in the Uranium cycle is a main concern for the produced radio- toxicity in the spent fuel and also for the proliferation resistance and the question will be discussed elsewhere during the meeting.

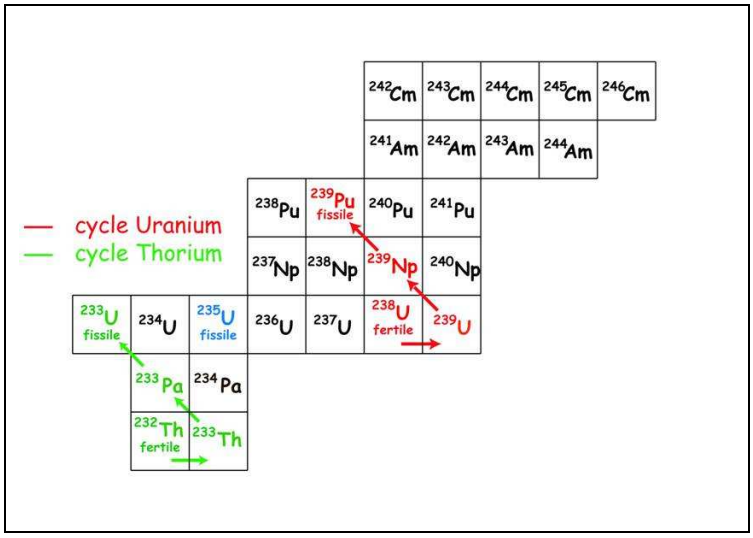

Figure 1 Actinide Chart

Natural Thorium does not contain fissile material and must therefore be mixed with fissile material produced elsewhere. So up to now it has only been used in experimental reactors fuelled with existing ^{235}U or Pu to extend the capabilities of the used fuel. For reasons that will soon become apparent, Thorium when compared to Uranium, has very interesting potential for energy production as well as nuclear waste minimization.

2. Comparisons between the Thorium and Uranium cycles.

The condition to obtain breeding in a fission reactor is that the number of nuclei that fission is smaller or at most equal to the number of fissile nuclei created by neutron capture on the fertile nuclei and subsequent decays during the same time span. If ν is the number of neutrons emitted by fission and α, the ratio of capture to fission cross-sections for a fissile nuclei as a function of energy, the number of neutrons available for breeding is given by $N_b = \nu-2(1+\alpha)$ and plotted figure 2 for the two fertile elements as a function of the neutron energy.

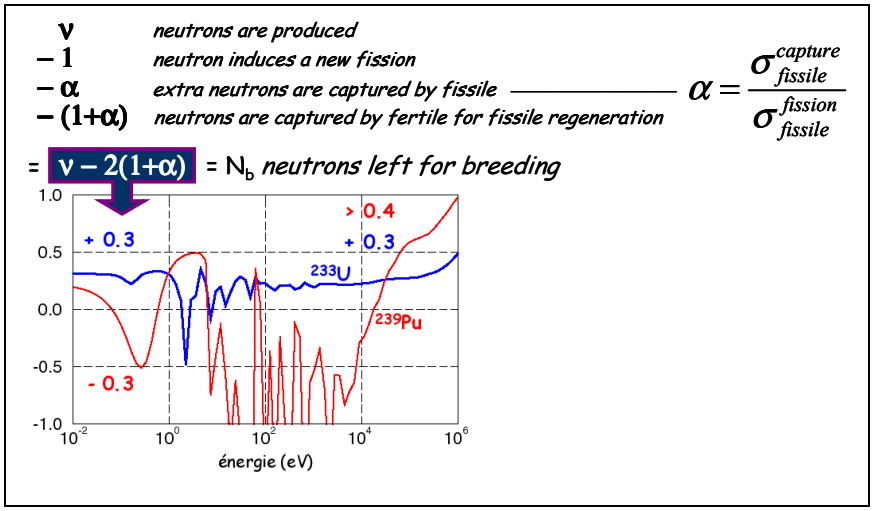

Figure 2 Available neutrons

As long as the available neutron number is always slightly larger than 0, breeding is possible. This is the case for the whole neutron energy spectrum for Uranium 233 whereas it is only possible for neutron energy larger than a few ten keV for Plutonium 239. This explains why, if Plutonium is produced and partly burnt in the light water reactors, it is impossible to reach an interesting breeding ratio with a thermal neutron spectrum. The main advantage of the thermal spectra is that the required fissile material for starting the chain reaction is smaller (factor up to six as we will see later) than for the fast neutron reactor. Another interesting feature of the Thorium cycle is the lower production of actinides which are the main contributors to the radio-toxicity of the spent fuel. Figure 1 shows that five successive neutron captures are necessary to reach Neptunium, whereas the Uranium cycle is already very close to the Minor Actinides. The radio-toxicity which tries to assess the risk due to the spent fuel of the various fuel cycles as a function of time is given on figure 3 which shows clearly the advantage of the Thorium cycle and we will come back to the actinides production later in the paper. As the number of available neutrons is always small ($N_b \sim 0.3$), it is very important to minimise all the potential neutron losses. As some fission products capture neutrons easily, it is also important to remove them as soon as possible from the reactor core and it is one of the reasons why the Thorium cycle has been linked to the molten salt reactors from the beginning.

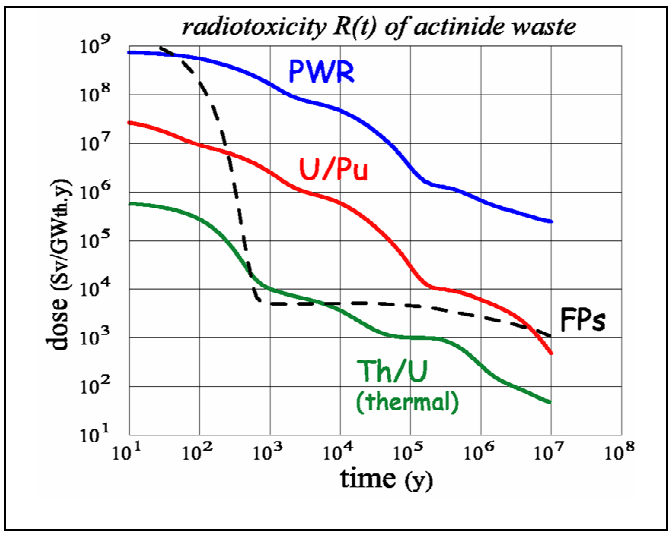

Figure 3 Radio-toxicity for the various cycles.

3. The MSBR project: review and discussion

MSR (Molten Salt Reactor) concepts were studied at first at the Oak Ridge National Laboratory (ORNL) with the ARE (Aircraft Reactor Experiment) [1] which was based on a liquid Uranium fluoride circulating in a BeO moderator and which ran for around a hundred hours. Studies were then oriented towards civilian applications such as electricity production. The Molten salt Reactor Experiment (MSRE) [2] managed from 1964 to 1969 the operation of a 8MWth graphite-moderated Molten Salt Reactor with a liquid fluoride fuel mixed with lithium and beryllium fluorides. Initially, the fuel was ^{235}Uranium, then ^{233}Uranium and finally Plutonium was also burnt. The main results were a very good operating performance for over four years, improvement of material against corrosion and a good understanding of fuel behaviour. These studies led ORNL to present the Molten Salt Breeder Reactor (MSBR) project [3] of a 1 GWe industrial reactor based on the Thorium cycle and sketched on figure 4.

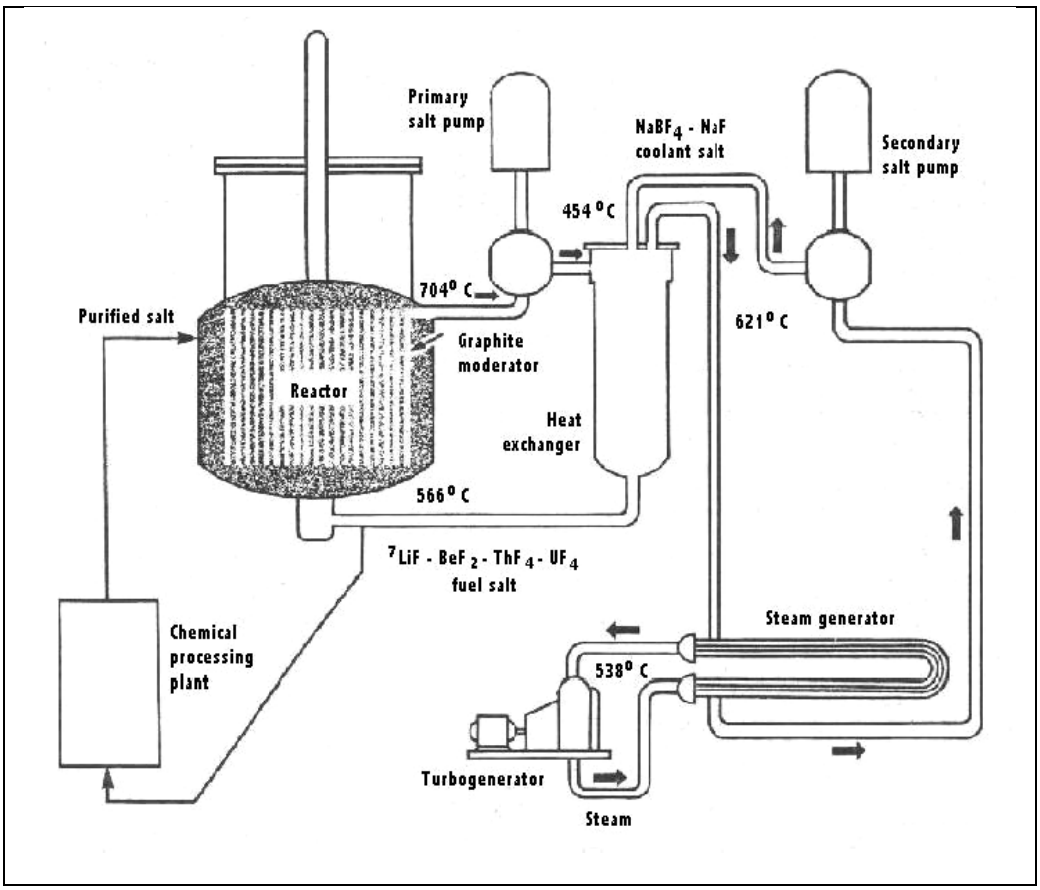

Figure 4. The MSBR project

The LiF-BeF_2-ThF_4 salt (~45 m^3) circulates through a graphite moderated core, pumps with a bubbling system and heat exchangers located near the core and partially through a pyro-chemical reprocessing unit. The bubbling system is running continuously and is assumed to extract continuously the gaseous and non soluble fission products which are the most neutron capturing. In the chemical unit which reprocess the whole salt volume within ten days, to obtain the largest breeding ratio, the Protactinium is quickly extracted with a full efficiency and temporarily stored to decay into ^{233}U which is re-injected in the core with all the actinides whereas the lanthanides are extracted with 20% efficiency and stored. The doubling time was calculated to be about 25 years for an ^{233}U initial loading slightly larger than one ton. At the end, the concept was not retained and ORNL studies were stopped in 1976. Some additional works have been made on the MSR concepts in France, Japan and Russia and since 2001 a review [4, 5] has been made in the EURATOM Concerted Action MOST. The last studies on MSBR had shown two things: the fast reprocessing scheme is somewhat impractical and may lead to too large Thorium losses and the global temperature feedback coefficient is positive due to positive graphite effects and makes the reactor not intrinsically safe. In 1999, we decided, in collaboration with EDF [6, 7], to revisit the MSBR concept from the point of view of the reprocessing constraints and of the intrinsic safety [8, 9]. The results presented here have been obtained with a stochastic code based on the neutron transport code MCNP [10] used with the ENDF/B-VI, JENDL 3.2 and JEF 2.2 data bases in that order to make feedback coefficients calculations and coupled with Bateman differential equations to make materials evolution calculations which allow us to know at each time the exact composition of the core. The evolution calculation is sketched on figure 5.

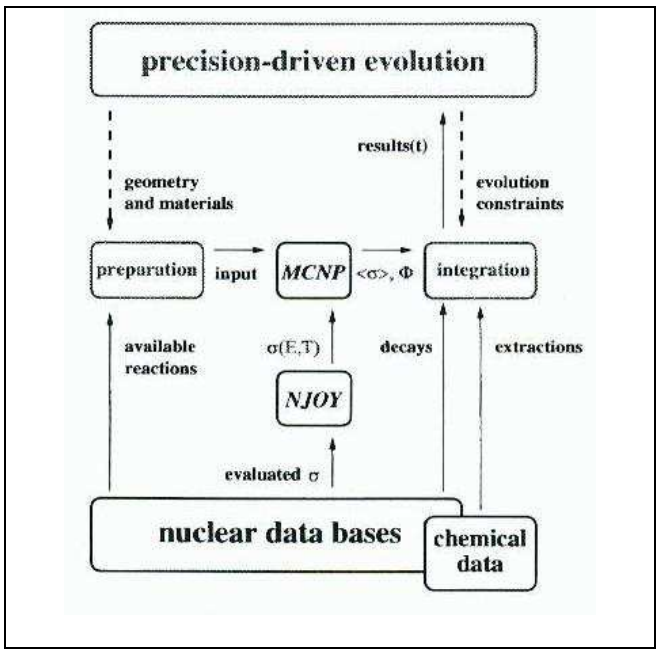

Figure 5 Organization of evolution tools around MCNP

4. The TMSR (Thorium Molten Salt Reactor) concepts.

The reference concept chosen for our studies in the continuation of the MSBR, is described now: The core is sketched on figure 6, it is a cylindrical assembly (1.6 m radius, 3.2 m high for the reference concept)) of graphite hexagons (15 cm side), each pierced by a channel of variable diameter which allows varying the moderation ratio during the studies.

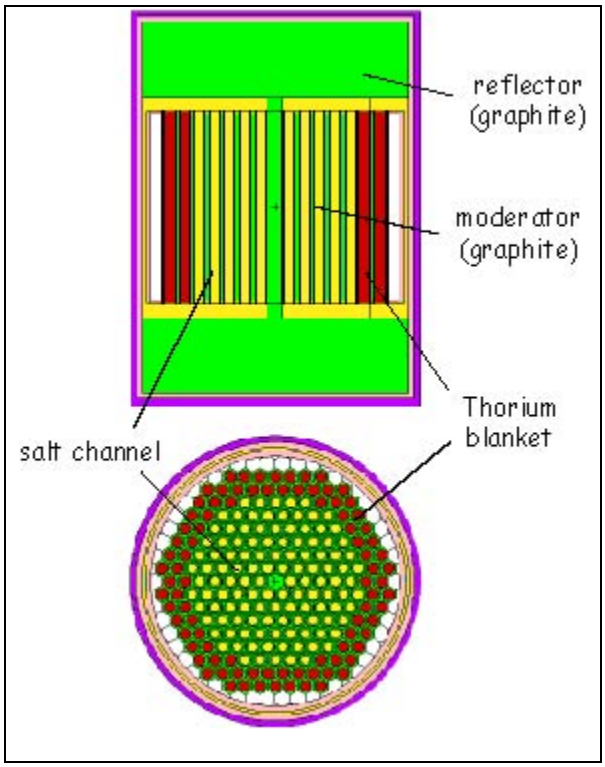

Figure 6: TMSR core representation

The graphite has a density of 1.86 g/cm3 and the salt is LiF (78%)- (HN) F_4 (22%) at 630°C near the eutectic point where HN stands, at the beginning, for Thorium and Uranium 233 in a quantity suitable to make the reactor critical. Then the produced actinides and fission products are taken into account in the neutron balance. The volume of the salt is equal to 20 m^3, whatever the concept, one third being out of the core. The core comprises also two plena above and under the graphite matrix and is surrounded by a radial reflector made with the same graphite hexagons hollowed out with cylinders with a radius of ten centimetres where a fertile salt with only Thorium as heavy nuclei is included to increase the ^{233}U production. This Uranium is also extracted every six months. Three concepts have been more extensively studied, one which is directly derived from the MSBR is the reference TMSR and the radius of the channels in the graphite is 8.5 cm (epithermal concept), the second one corresponds to the case where there is no graphite inside the core and the reflectors are made of non moderating materials (fast concept) and the third one is the thermal concept where, to obtain acceptable reactivity feedback coefficients, the hexagon sides have been reduced to 5cm and the salt channel radii to 1.33 cm. The geometrical characteristics are summed up in table 1

Table 1. Description of the various concepts

	MSBR	Thermal TMSR	Epithermal TMSR	Fast TMSR
Hexagon side(cm)	15.0	5.	15.	-
Channel radius (cm)	7.5	1.33	8.5	-
Core radius (m)	2.3	2.55	1.6	1.25
Core height (m)	4.6	5.3	3.2	2.6

The second modification affects the reprocessing scheme, the new one is shown on figure 7. The only continuous action is the bubbling which is assumed to extract within 30 s., the gaseous and non soluble fission products present in the salt. The salt properties are monitored on line and in addition we consider a delayed and separated reprocessing of the whole salt over a six months period. Uranium (including ^{233}U) is extracted by fluorination and directly and immediately reintroduced in the core. The other processes aim at the lanthanides extraction which may require previous the TRU and Thorium extraction and are made in a near but separate chemical unit.

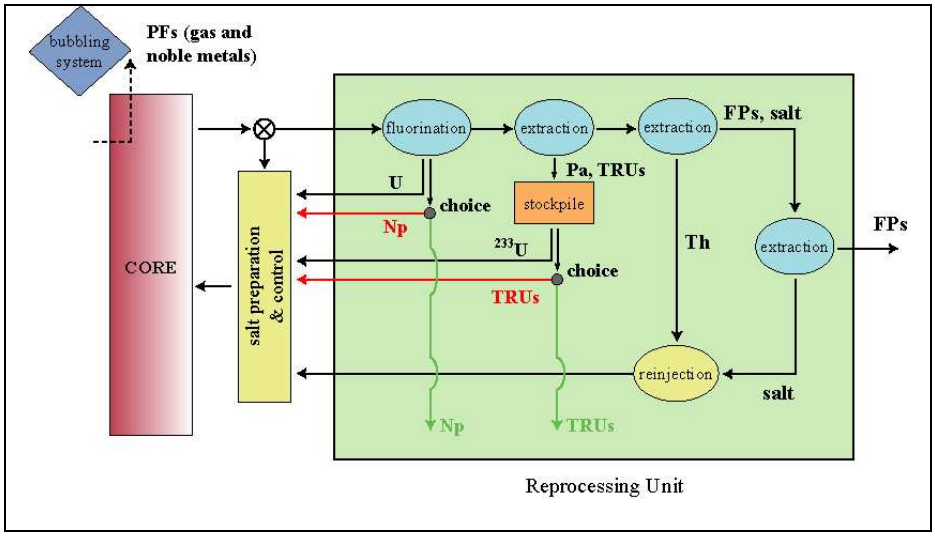

Figure 7 Reprocessing scheme

The possibility to send the Pa and the TRU back into the core or not leads to different inventories at equilibrium which will be given later. For the three concepts we have checked that the global reactivity coefficients are negative (see table 2) that ensures an inherent safety for the reactor. The calculations have been made independently for the whole reactor, for the graphite and for the salt taking into account the two main effects, the Doppler and the density effects.

Table 2. Reactivity coefficients

	MSBR	Thermal TMSR	Epithermal TMSR	Fast TMSR
Doppler effect	- 3.3	- 3.2	- 6.1	- 3.2
Salt density effect	+2.4	+0.6	+3.2	- 2.2
Graphite effect	+1.6	+1.8	+0.5	0
Global effect(pcm/°)	+0.7	- 0.8	- 2.4	- 5.4

The sum of the reactivity coefficients for the salt alone is always negative, that is a pledge for the prompt safety. More detailed calculations, taking into account the salt circulation and the temperature differences, are underway to check more carefully the molten salt reactor safety. But at that time, with these calculated negative reactivity coefficients, with the non appearance of pressure even at high temperature, with the absence of reactivity reserve and with the possibility to drain the salt quickly in safety tanks, the Thorium molten salt reactors appear as very attractive when considering the safety point of view.

We will now give the results of the calculations concerning the production of the various actinides during the reactor operation and we will discuss these results from the point of view of the reduction of waste production and of the proliferation concern.

5. Actinide yields

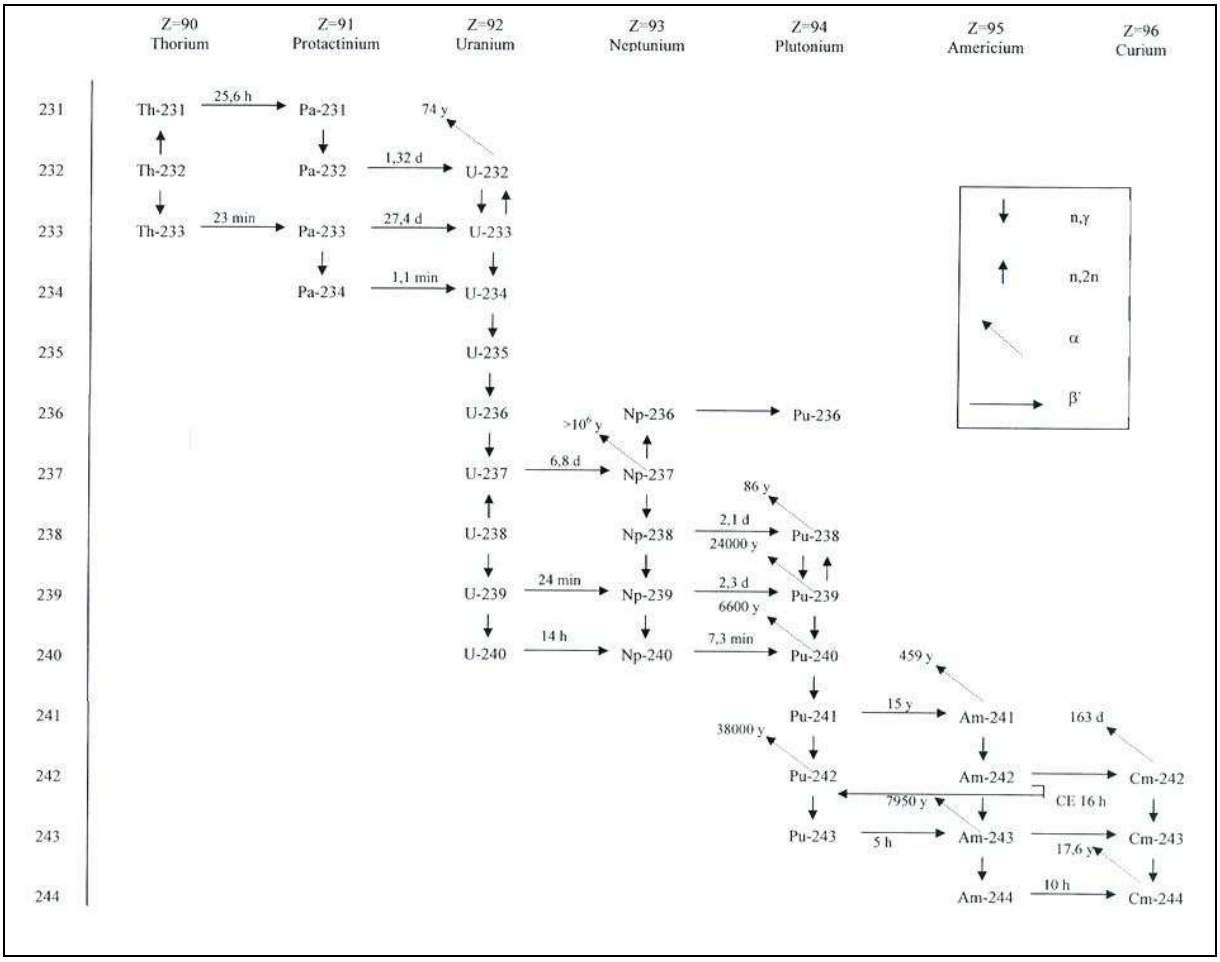

Figure 8. Thorium and Uranium chains

The Thorium chain is shown in the upper part of figure 8 and we see clearly that the transition to the Uranium chain (figure 8 lower part) is made through the ^{235}U, a fissile nucleus and that the ^{237}Np, first Minor Actinide found in the diagram, is produced after five neutron captures and three β-decays.

The occurrence of that processes is mainly dependant of the neutron spectra and fluxes, so the results will be presented in various conditions up to the equilibrium which, in some cases, would require to run during one hundred years and even more. This is illustrated on figure 9 where the quantity of the various actinides and Uranium isotopes present in the core of a MSBR-like reactor is given as a function of the running time.

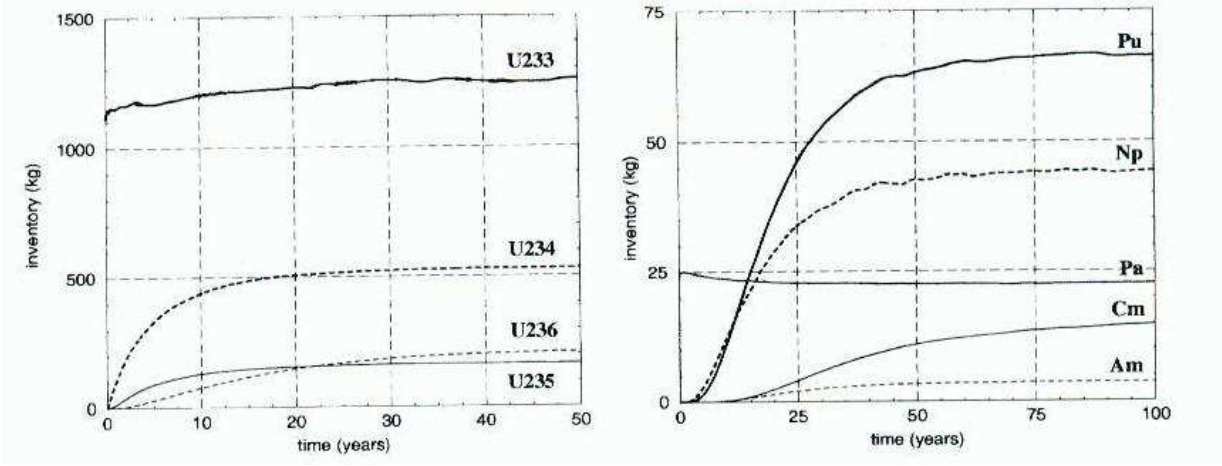

Figure 9. Uranium (left) and actinides (right) production in the MSBR

Even if equilibrium is not clearly reached for Curium and Americium after one hundred years, we will take the obtained values at that time to compare them in various situations. The values obtained for the MSBR and the TMSR under various conditions are given on table 3 in the same running assumptions, which is that the actinides, if separated, are sent again into the core after their extraction, that explains the slow build-up of the higher mass actinides.

Table 3. Actinide inventory at equilibrium (kg) for closed cycle

	MSBR	Thermal TMSR	Epithermal TMSR	Fast TMSR
Th	65 560.	48 400.	45 300.	43 300.
Pa	22.5	75.	74.	85.
U	2 156.	1 600.	4 200.	8 300.
Np	44.0	29.	110.	150.
Pu	66.	38.	260.	270.
Am	3.6	3.1	7.1	4.8
Cm	15.1	14.1	18.0	2.4
Bk	0.01	0.005	0.04	0.0002 (100y)
Cf	0.081	0.06	0.11	0.0007 (100y)

The obtained values are very low especially for the heaviest Minor Actinides and illustrate very well the interest of the Thorium cycle for minimisation of long life nuclear waste production The actinide amount sent to the wastes will be proportional to the actinide quantity present in the core and to the efficiency of the lanthanides extraction process which is not chosen now but in any case will be very small. The Minor Actinide production is compared in table 4 with the values calculated for the Light Water Reactors and the Fast Neutron Reactors [11] for a 1Gwe (~7 TWhe each year). The table gives the inventories which have to be taken in charge when the reactor is stopped. The interest of the MSR in Thorium cycle appears clearly as the only comparable values are in the Neptunium isotopes, the

other actinide values being significantly lower. Here is the explanation of the radio-toxicity curves presented on figure 3.

Table 4. Comparison between various nuclear reactor concepts in closed fuel cycles. Inventories (kg)

	Thermal TMSR	Epithermal TMSR	Fast TMSR	FNR	PWR
Np	29.	110.	150.	70.	91.5
Pu	38.	260.	270.	12 550.	3 850.
Am	3.1	7.1	4.8	528.	248.
Cm	14.1	18.	2.4	135.	124.

The results emphasize clearly the interest of the Thorium cycle for minimisation of the heaviest Actinides (Pu and heaviest) which are the major contributor to the radio-toxicity of the nuclear wastes. Only the Fission Products (less than one ton each year) have to be managed and moved elsewhere.

Another way of running might be to burn the extracted TRU in separate reactors which would be dedicated to actinides (mainly Neptunium and Plutonium) burning, in that case the M. A. inventories are clearly lower and the calculation leads to the values given in table 5 where the quantities of actinides present in the core at equilibrium are compared again with other reactor cycles. The results are still very appealing, the values that have to be taken into account each year being very low and the quantities extracted to be burnt elsewhere each year rather limited.

Table 5. Comparison between various nuclear reactor concepts in open cycles. Inventories (kg)

	Thermal TMSR	Epithermal TMSR	Fast TMSR	FNR	PWR
Np	7.0	15.0	9.7	23.0	102.
Pu	1.9	2.8	0.6	12 250.	1 420.
Am	$4.0\ 10^{-4}$	$5.0\ 10^{-4}$	$7.0\ 10^{-7}$	192.	86.0
Cm	0.001	$1.0\ 10^{-4}$	$2.0\ 10^{-8}$	15.0	14.0

As a partial conclusion, we observe that the actinides quantity in the core is very small even for Np and Pu, whatever the chosen hypothesis and that the radioactive materials quantity that have to be sent out of the nuclear plant is also very small.

The tritium production is often considered to be important in the MSR, so we have calculated the production for the studied concepts. The main source is the ^{6}Li through the reaction:

$$n + {}^{6}Li \longrightarrow t + \alpha$$

The Lithium 6 comes at the beginning from the non separated part in the initial Lithium which will disappear within the five first running years and in the case of the presence of beryllium in the salt, ^{6}Li is permanently produced through the reaction

$$n + {}^{9}Be \longrightarrow {}^{6}He + \alpha,$$ ^{6}He decaying quickly in ^{6}Li.

The second reaction producing tritium is directly due to the ^{7}Li and the cross section is smaller than for the ^{6}Li.

$$n + {}^{7}Li \longrightarrow t + \alpha + n$$

The produced quantity, in the case of the MSBR, amounts to 0.385kg/year at the beginning and decreases to 0.150 kg/year at equilibrium after five years; in the case of the TMSR concepts, the tritium production begins at the 0.185 kg/year level and equilibrates at 0.11kg/year. The values are non negligible but, for the TMSR, are always smaller than the 0.280kg/year obtained in the Candu reactors now at work.

6. Non proliferation considerations

Our reactor is working with ^{233}U which, due to its small critical mass, around 15 kg, and its half life, 1.5 10^{5} years, is an interesting potential material with which to make nuclear weapons. We will therefore examine the Uranium cycle during the whole reactor operation. The Uranium fuel is diluted in the salt and represents a few percent in mass of the salt. So to obtain the Uranium quantity sufficient to make a weapon, requires a chemical unit able to process at least a few tons of salt. As the salt composition is continuously monitored, there is no reactivity reserve in the core. Moreover as we have seen in the preceding chapter, the various Uranium isotopes are quickly produced and are mixed with the ^{233}U and are extracted together in chemical processes such as fluorination. The build-up of Uranium isotopes is shown in figure10 and the Uranium isotope production values are given on table 6 for the various concepts presented here. Their presence increases the critical mass and then the requested salt quantity to be treated to obtain it and that is equivalent to an isotope dilution. The only way to obtain pure ^{233}U is to use an efficient and fast Protactinium separation and to let it decay out of the neutron flux. It was the case for the MSBR project which could produce 38 kg each year; in the TMSR cases, there is no ^{233}U available because the regeneration is obtained from the Uranium produced, not only in the core but also in the axial blanket from where it is extracted every six months.

Table 6. Uranium isotope inventories at equilibrium (kg)

	MSBR	Thermal TMSR	Epithermal TMSR	Fast TMSR
^{232}U	2.8	2.5	3.7	14.0
^{233}U	1 250.	790.	2 400.	5 200
^{234}U	530.	470.	1 100.	1 900.
^{jk235}U	160.	100.	410.	560.
^{236}U	210.	250.	380.	580.
^{238}U	2.8	4.8	1.5	0.7
Total U	2 155.6	1 616.	4 290.	8 254.

Concerning proliferation resistance, the most interesting product is Uranium 232 which is mainly produced (see figure 8) by a (n, 2n) reaction on Thorium and to a lesser extent by a (n, 2n) reaction on ^{233}U. These reactions have a high energy threshold but figure 10 shows that, whatever the concept, the production of ^{232}U will still occur. On the left are given as a function of the neutron energy, the cross-

sections for the (n, 2n) reactions and the neutron fluxes in the core and the blanket for the various reactor concepts; on the right, is presented the quantity of ^{232}U related to the whole Uranium present in the salt as a function of the graphite channel radii, that is to say as a function of the moderation ratio for the neutron flux. Contrary to the results given table 6, the ^{232}U production given on the right part of figure 10 are calculated with the hypothesis that the ^{231}Pa was not sent into the core after separation and ^{233}Pa decay. This minimizes by a factor 2.(thermal concept) to 7.(fast concept) the ^{232}U production and we are sure even in that case that the produced quantity is enough to prevent ^{233}U diversion.

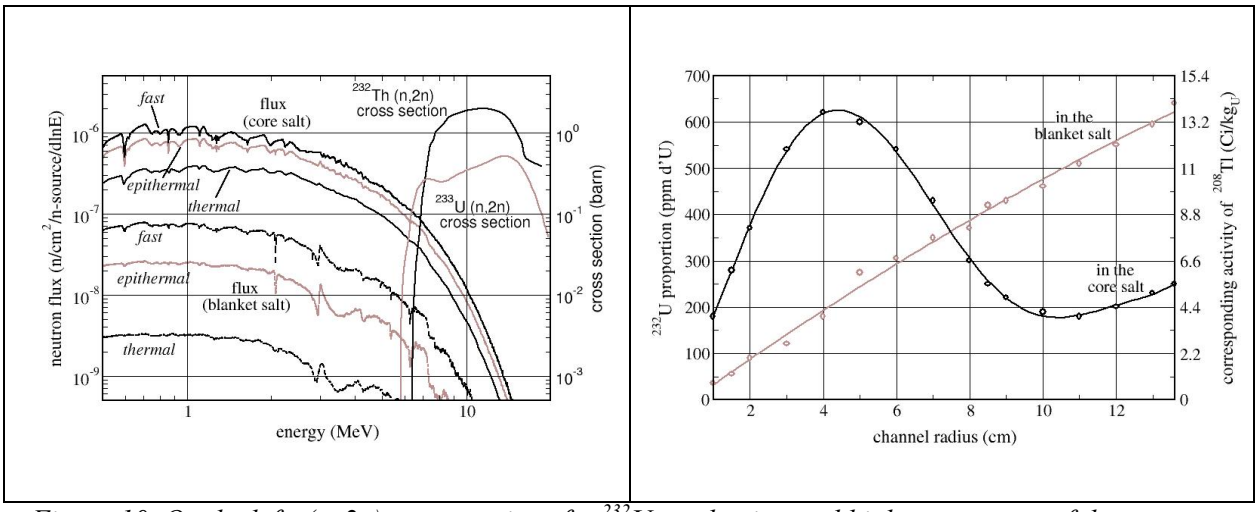

Figure 10. On the left: (n, 2n) cross sections for ^{232}U production and high energy part of the neutron flux for the various concepts. On the right: proportion of ^{232}U in the Uranium present in the core as a function of the moderation ratio.

The proportion is also given for the Thorium salt in the reflector and even in that case the ^{232}U production is not at all negligible. Another interesting result is the weak sensitivity to the neutron spectrum which implies that, whatever the initial load and running condition of the reactor, the inventory will contain a noticeable amount of ^{232}U. The decay scheme of ^{232}U (half life 68.9 years) is given on figure 11.

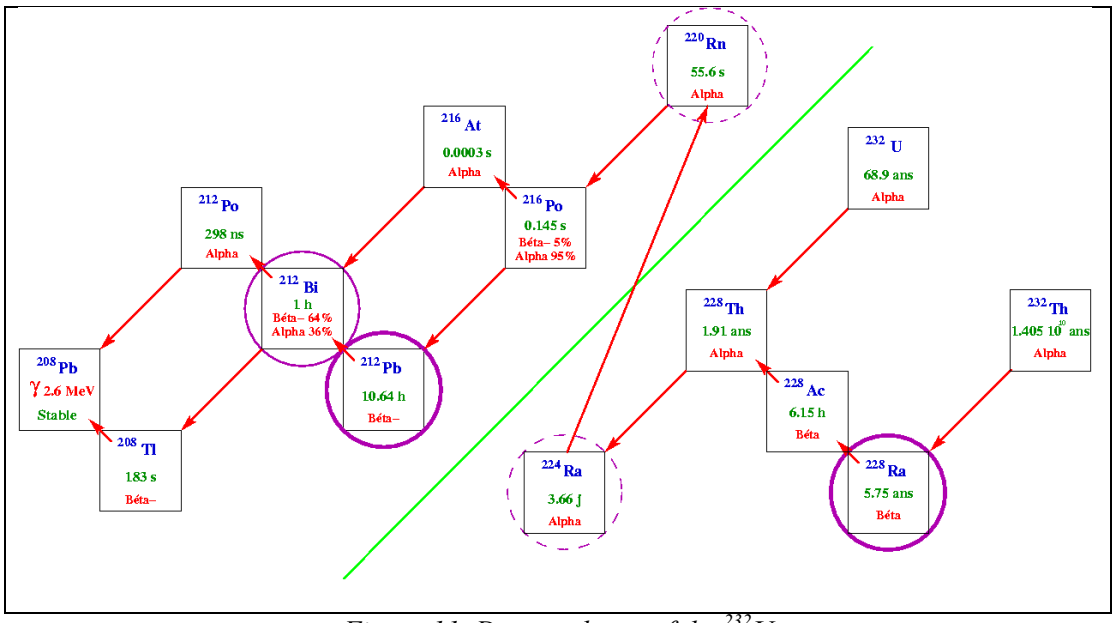

Figure 11. Decay scheme of the ^{232}U

The main feature related to the proliferation resistance is the presence in 36% of the ^{232}U decays of a very energetic (2.6 MeV) γ ray which prevents easy manipulation of the salt and above all of the extracted Uranium and which may therefore help to detect the diversion of Uranium even in small quantities. The slowest step in the decay chain is the ^{228}Th decay (1.91 y). The activity related only to that γ ray, assuming equilibrium between the descendants, is 250 GBq for 1 kg of extracted Uranium with a ratio $^{232}U/U$ of 250 ppm. That value combined with the γ energy explains why the manipulation and the transport of diverted Uranium are virtually impossible without their detection and present a serious hazard to their transporters. An illustration of the activity growth is given on figure 12 which is extracted from the reference [12].

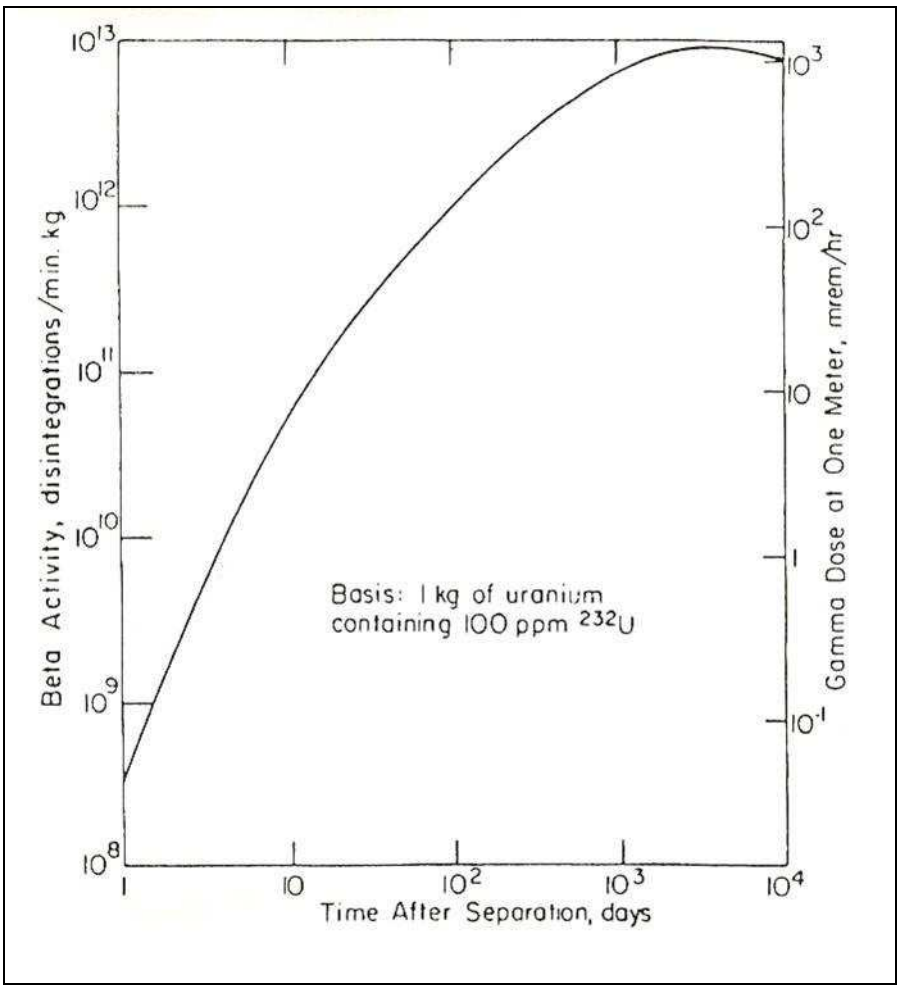

Figure 12. Growth of the activity [12]

The weakest point of the MSBR project, where the Uranium balance was favourable and where the Protactinium was quickly extracted and efficiently separated to let the ^{233}Pa decay in ^{233}U, is the possibility to divert some part of that Uranium at the right time to obtain rather pure ^{233}U. The TMSR project is calculated to run without the capability of producing extra Uranium, the reprocessing unit (figure 7) aim is to extract the lanthanides but before that extraction, it may be necessary to extract before, TRU, Pa and Thorium. If the extracted Pa, TRU and Thorium are sent again directly in the core, there is no problem; in the other case, it would be necessary to leave at the first fluorination a sufficient percentage of Uranium which will be mixed to the Uranium coming from the Pa decay to obtain Uranium with enough ^{232}U.

Another interesting TMSR feature is that the fast neutron spectrum option may run during 20 years without salt reprocessing, that is to say without reprocessing unit but that option requires around 5.5 tons of ^{233}Uto start the chain reaction. Some possibilities exist to decrease this quantity and have been presented in [13]. With the reprocessing unit in the vicinity of the reactor, the problems related to the fuel transport are greatly reduced and this unit may be adapted at the beginning to receive the Thorium irradiated in other reactors to produce the first U3 load without any other manipulations. As the chemical reprocessing schemes are not firmly established now, it is difficult to go further in definite conclusions about the proliferation issues concerning the molten salt reactors in Thorium cycle. But it is clear that the unavoidable production of the Uranium232 together with the Uranium 233 production is the main obstacle because its presence prevents easy manipulations and transport of the fissile material.

From the point of view of proliferation, the Plutonium has been taken into account and does not matter because it is produced in very limited quantity and the larger part (more than 60%) is ^{238}Pu which is characterized by a large heat release; so MSRs are not convenient at all to make nuclear weapons with the produced Plutonium.

7. Summary

The Thorium cycle presents very interesting characteristics for nuclear energy production. It has been demonstrated that it is possible to obtain a breeding ratio at least equal to one with any kind of neutron spectra in the reactor and to obtain good reactivity feedback coefficients ensuring inherent safety conditions. Therefore the reactor is well suited to the objectives that have been defined for the future nuclear reactors by the Generation IV international forum. As shown in the paper, from the physical point of view, the Thorium molten salt reactors allow us to satisfy the main criteria now requested for a sustainable nuclear energy production: resource saving, intrinsic safety, waste production minimisation, no reactivity reserves and good resistance to proliferation. A lot of possibilities have been presented which need further studies according to the priorities and technologies that will be chosen. But in any case the Minor Actinides production is very low and that will simplify greatly the management of the wastes which will mostly consist of fission products. In the MSR, the fissile material is disseminated in small quantity (1- 3%) in the salt and requires reprocessing a large amount of salt to obtain sufficient quantity of fissile material. Moreover, the unavoidable production of ^{232}U accompanying the ^{233}U production, which might be a large problem in the case of solid fuel preparation gives very strong constraints on the manipulation of the Uranium and helps prevent undesirable use and transportation.

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