CRITICAL AND SUB-CRITICAL GT-MHRs FOR WASTE DISPOSAL AND PROLIFERATION-RESISTANT FUEL CYCLES

D. Ridikas, G. Fioni, P. Goberis, O. D'eruelle, M. Fadil, F. Marie
CEA Saclay
91191 Gif-sur-Yvette Cedex, France

Abstract

The gas turbine modular helium-cooled reactor (GT-MHR), using an annular graphite core and graphite inner and outer reflectors, is known probably as the best option for the maximum plutonium destruction in once-through cycle. Combination of the critical GT-MHR with an accelerator driven (AD) sub-critical GT-MHR core (AD-GT-MHR) would allow to merge the best features of both systems to achieve near total destruction of fissionable plutonium. We perform detailed simulations along these lines and compare different scenarios in order not only to reduce considerably the mass of plutonium isotopes but also to diminish the waste radiotoxicity in the long term.

1. Introduction

Nuclear waste radiotoxicity in the long term, say, more than 1 000 years, is strongly dominated by plutonium isotopes if the spent fuel originating from the once-through cycle is considered as waste. The disposal of this waste has become an environmental and political issue. The main concerns are related to the potential for radiation release and exposure from the waste, and also the possible diversion of fissionable material.

Plutonium is the unique waste component. It is fissionable, and therefore capable of releasing significant amount of energy. It is also a hazardous material, particularly if inhaled in particulate form. Because of its potential use in nuclear weapons, there is great sensitivity about isolating plutonium from other components of the nuclear waste stream.

As it was shown in a number of studies, gas-cooled reactor technologies offer significant advantages in accomplishing the transmutation of plutonium isotopes and nearly total destruction of ^{239}Pu in particular (see [1-7] and also references therein). GT-MHR uses well-thermalised neutron spectrum, operates at high temperature without the need for fertile material and employs ceramic-coated fuel. It utilises natural erbium as a burnable poison with the capture cross-section having a resonance at a neutron energy such that ensures a strong negative temperature coefficient of reactivity. The lack of interaction of neutrons with coolant (helium gas) means that temperature feedback is the only significant contributor to the power coefficient. As a matter of fact, no additional plutonium is produced during the fuel cycle since no ^{238}U is used.

This paper describes the application of critical as well as sub-critical (accelerator driven) GT-MHR for transmutation of plutonium originating from the spent nuclear fuel in the once-through cycle. We examine a few different scenarios in order to obtain the maximum destruction of plutonium as well as to minimise the irradiated fuel radiotoxicity in the long term. MCNPX [8], MCNP4B [9], MONTEBURNS [10] and CINDER'90 [11] codes are employed at different stages of our simulations. The performances of the codes have been reasonably benchmarked in part in [12] by simulating the fuel cycle of the high flux reactor at ILL Grenoble.

This work is organised as follows. Major GT-MHR characteristics and system modelling details are summarised in the following two sections. After we present different scenarios considered, which are followed by simulation results and discussion. The paper ends with conclusions and outlook.

2. Major GT-MHR characteristics

The Gas Turbine – Modular Helium Reactor (GT-MHR) [1] is an electric generation power plant that couples the passively safe critical reactor with a highly efficient energy conversion system. Conceptual design of GT-MHR was developed in a joint project of the Russian Federation, USA, France and Japan with the major interest in plutonium based fuel cycles in general and weapons grade plutonium (WGPu) destruction in particular. We refer the reader to Reference [1] for further details on technical characteristics of GT-MHR. Table 1 gives only major reactor parameters essential for the modelling of the system.

A simplified model of GT-MHR reactor which is shown in Figure 1 has been created using MCNP4B geometry set-up [9]. It consists of the reactor core (C), inner reflector (Ri), side reflector (Rs), top reflector (Rt), bottom reflector (Rb), and reactor vessel (V) as shown explicitly in Figure 1. Further details on modelling can be found in [13], while some of the results in part have already been reported in [14].

Table 1. **Basic GT-MHR reactor parameters**

Power, MW(th)	600
Active core size:	
- height, cm - outer diameter, cm - inner diameter, cm	800 484 296
Active core volume, m^3	92.1
Average power density, W/cm^3	6.5
Outer diameter of the side reflector, cm	684
Total height of the core, cm (with top and bottom reflectors)	1 090
Vessel size:	
- height, cm - outer diameter, cm - thickness, cm	2 022 750 20
Averaged temperature, °C:	
- active core - inner reflector - side reflector - top and bottom reflector - vessel - helium at the core inlet/outlet	1 230 1 500 1 100 1 000 440 487.8/850.0

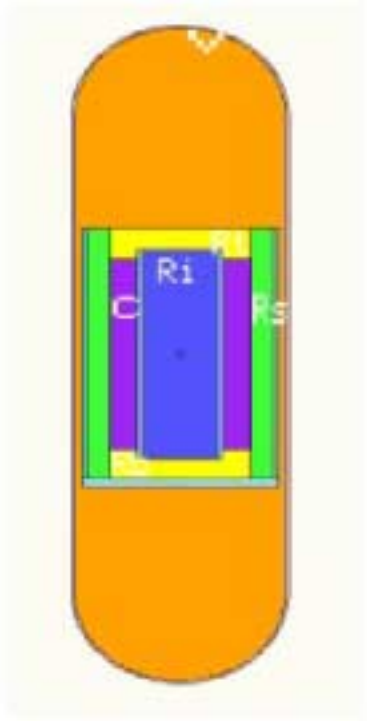
3. Modelling tools and procedure

The major reactor parameters used for modelling of the GT-MHR reactor are given in Table 1. As we have already mentioned above, a three-dimensional geometry set-up was constructed and is presented in Figure 1. MCNP4B was also used to obtain k-eigenvalues and neutron fluxes. In the case when a sub-critical GT-MHR (AD-GT-MHR) is considered, a single difference was the prevision for an accelerator target (for neutron production by spallation) and surrounded pressure vessel, both located in the central graphite reflector. We have considered a fluidised bed of tungsten particles cooled by helium as a spallation target. In this particular case the MCNPX code in proton source mode was applied to all calculations of neutron fluxes.

Burn-up calculations were made with MONTEBURNS [10]. MCNPX [8] was used to write a low energy (E_n<20 MeV) neutron source in the spallation target, that was employed for the burn-up calculations with MONTEBURNS. Corresponding activities and radiotoxicities were calculated with CINDER'90 code system [11].

As soon as k_{eff}<1 the length of the fuel cycle could be determined in effective days at full power in the case of a critical system. For a sub-critical system k_{eff} was "allowed" to vary within certain interval where neutron multiplication could be still compensated by a proton accelerator, say, at maximum ~60 MW of beam power of 1 GeV protons. If this condition was not fulfilled any longer, the total core power was decreased accordingly with decreasing k_{eff}.

Figure 1. **Lengthwise section view of GT-MHR reactor model.**
The following notation is employed: C – core, Rt – top reflector,
Ri – inner reflector, Rb – bottom reflector, Rs – side reflector, and V – reactor vessel.

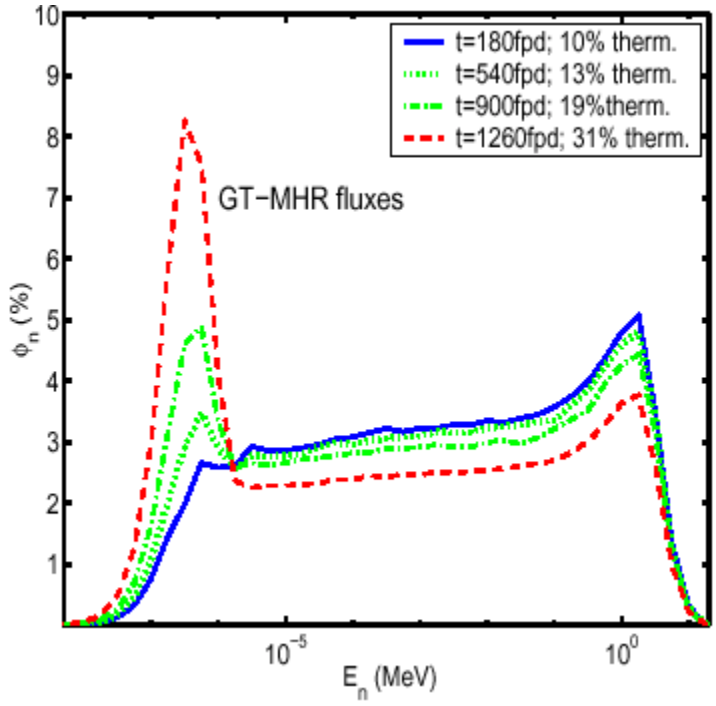


The change of neutron flux spectra because of fuel burn-up also has been determined. A typical neutron spectrum evolution for plutonium fuel poisoned with natural erbium is shown in Figure 2. The observed increase of the thermal flux can be explained by the loss of ^{239}Pu and ^{240}Pu in addition to burnable poison ^{167}Er during the fuel burn-up. In other words, the mass of the elements with the highest thermal neutron capture cross-sections is considerably decreased with time.

We note separately at this point that the change of the energy spectra of neutrons will change the average cross-sections to be used in the burn-up calculations, in some cases by a factor of two or more [13,14]. Therefore for these types of spectra fuel evolution calculations have to be performed with corresponding variable neutron fluxes as it is done with MONTEBURNS [10].

Another important result of our calculations is related to the estimation of neutron fluxes in their absolute value, again as a function of time. For example, we found that typical average GT-MHR neutron fluxes in the active core may increase by 50-100%, i.e. from ~$1\cdot10^{14}$ n/(cm^{2}s) to ~$2\cdot10^{14}$ n/(cm^{2}s) at the beginning and at the end of the fuel cycle respectively. This shows that irradiation/burn-up conditions may change as a function of time what has to be taken into account for the follow up burn-up calculations.

Figure 2. **Typical change of the averaged energy spectra of neutrons in the active core of GT-MHR for different fuel burn-up expressed in full power days (fpd).**



4. Scenarios examined

First of all, in Table 2 we fix the transuranic waste composition to be transmuted. This table represents typical form of LWR discharge (40 GWd/tonne burn-up) when uranium and fission products are separated. 1 500 kg of the waste is taken arbitrarily.

Table 2. **Initial composition of the TRU considered in this study.**

Nuclide	**(%)**	**(kg)**
^{237}Np	4.9	73.5
^{238}Pu	1.7	25.5
^{239}Pu	54.5	817.5
^{240}Pu	22.8	342.0
^{241}Pu	5.4	81.0
^{242}Pu	3.7	55.5
^{241}Am	5.7	85.5
^{243}Am	0.9	13.5
^{242}Cm	0.1	1.5
^{244}Cm	0.3	4.5
Total Pu	88.1	1321.5
Total 100	1 500	

Scenario S0. This is our reference point when TRU represented in Table 2 is left to decay naturally, i.e. no irradiation takes place.

Scenario S1. In this case only plutonium isotopes are placed in the critical GT-MHR for destruction. The length of the fuel cycle is ~1 550 days. Table 3 lists in detail initial load and discharge isotopic composition (see column Discharge S1) correspondingly. Total neutron fluence was ~$1.7 \cdot 10^{22}$ n/cm^2.

Table 3. **Isotopic composition of the TRU considered for transmutation: cases S1 and S2. 19.2 kg of ^{167}Er was put at the beginning of the fuel cycle for Scenario S1 and additional 13.5 kg of ^{167}Er was put at the beginning of the fuel cycle for Scenario S2 (at the end of the fuel cycle of Scenario S1).**

Nuclide	Load (kg)	Discharge S1 (kg)	Discharge S2 (kg)
^{234}U	0.0	0.6	0.6
^{235}U	0.0	0.2	0.2
^{236}U	0.0	0.1	0.1
^{237}Np	0.0	0.0	0.0
^{238}Pu	25.5	26.5	20.7
^{239}Pu	817.5	24.8	4.4
^{240}Pu	342.0	47.3	17.6
^{241}Pu	81.0	87.1	14.0
^{242}Pu	55.5	141.0	149.0
^{241}Am	0.0	9.1	2.1
^{243}Am	0.0	35.0	38.8
^{242}Cm	0.0	6.1	4.7
^{243}Cm	0.0	0.2	0.2
^{244}Cm	0.0	32.7	48.7
^{245}Cm	0.0	2.1	1.5
^{246}Cm	0.0	0.5	1.3
Total Pu	1321.5	326.5	205.7
Total	1321.5	<414	<305

Scenario S2. This case is actually a continuation of Scenario 1. Now GT-MHR is coupled to proton accelerator and run in its sub-critical mode for another 300 days with decreasing k_{eff} (k_{eff} ~0.90→0.52) and consequently decreasing reactor power P_{th} ($P_{th} = 1.0P_0 \rightarrow 0.25P_0$). The accelerator power should be increased from ~33 MW to ~66 MW. A detailed composition of TRU is presented in Table 3 (column Discharge S2). Total neutron fluence (including Scenario S1) was ~$2.2 \cdot 10^{22}$ n/cm^2.

Scenario S3. In this case GT-MHR is coupled to the proton accelerator at the very beginning since full TRU composition is selected including isotopes of Np, Am and Cm as shown in Table 4. System starts with k_{eff} ~0.91 and runs for ~500 days with decreasing accelerator power until the core becomes critical. At this moment there is no need for accelerator, and system runs in its critical mode for another ~900 days. Finally, GT-MHR finishes its cycle as a sub-critical system after additional ~200 days of operation with final k_{eff} ~0.88. Resulting TRU composition at the discharge is shown in fluence was ~$2.2 \cdot 10^{22}$ n/cm^2.

Scenario S4. This case is actually a continuation of Scenario 3. GT-MHR continues running in its sub-critical mode for another 250 days with decreasing k_{eff} (k_{eff} ~0.88→0.60) and consequently decreasing reactor power P_{th} ($P_{th} = 1.0P_0 \rightarrow 0.30P_0$). The accelerator power should be increased from ~42 MW to ~62 MW. A detailed composition of TRU is presented in Table 4 (column Discharge S4). Total neutron fluence (including Scenario S3) was ~$2.5 \cdot 10^{22}$ n/cm^2.

Table 4. **Isotopic composition of the TRU considered for transmutation: cases S3 and S4. 29.2 kg of ^{167}Er was put at the beginning of the fuel cycle.**

Nuclide	Load (kg)	Discharge S3 (kg)	Discharge S4 (kg)
^{234}U	0.0	1.7	1.7
^{235}U	0.0	0.4	0.4
^{236}U	0.0	0.1	0.2
^{237}Np	73.5	29.5	20.7
^{238}Pu	25.5	90.6	69.1
^{239}Pu	817.5	27.5	13.0
^{240}Pu	342.0	42.4	20.9
^{241}Pu	81.0	72.0	18.1
^{242}Pu	55.5	151.0	153.0
^{241}Am	85.5	11.9	2.9
^{243}Am	13.5	40.4	43.0
^{242}Cm	1.5	11.4	9.7
^{243}Cm	0.0	0.6	0.4
^{244}Cm	4.5	50.6	67.5
^{245}Cm	0.0	3.8	2.7
^{246}Cm	0.0	1.3	2.5
Total Pu	1 321.5	383.5	274.1
Total	1 500	<536	<427

5. Discussion

Tables 3 and 4 contain the burn-up results obtained for 4 different scenarios considered. What should be said at the very beginning is that ^{240}Pu appears as a fertile material no matter which scenario is chosen.

Discharge S1 in Table 3 simply shows that very high burn-up rates can be reached even if only a critical GT-MHR is considered: ~97% for ^{239}Pu and ~75% for all Pu isotopes. Indeed, coupling of the critical GT-MHR to an accelerator gives an extremely deep burn-up, ~99.5% for ^{239}Pu and ~84.4% for all Pu isotopes. These calculations confirm earlier estimates of the similar type reported in [4,6].

It should be noted separately that after Scenario S1 there was no real decrease in the mass of ^{241}Pu, simply because its destruction was compensated by its creation from ^{240}Pu. Therefore, Scenario S2 is essential not only for deep burning of ^{239}Pu, but also for considerable elimination of equally proliferation-offensive ^{241}Pu.

Scenario 3 shows that mixing of waste plutonium with the rest of actinides (Np, Am, Cm) can influence considerably GT-MHR performances. First of all, the system cannot start its cycle in the critical mode, i.e. an accelerator is needed already at the very beginning of the operation. Secondly, as soon as the sub-critical GT-MHR starts its fuel cycle, k_{eff} is constantly increasing as a function of time; the system becomes critical and can run in its critical mode for a certain period of time without accelerator. At this point there is a saturation of the ^{241}Pu build up from ^{240}Pu, i.e. until now ^{241}Pu mass was constantly increasing and starts decreasing from now on. Finally, due to considerable burn-up of ^{239}Pu (compared to its initial value) and also ^{241}Pu (compared to its saturation value) k_{eff} falls below unity and there is again need for an accelerator to continue the fuel cycle.

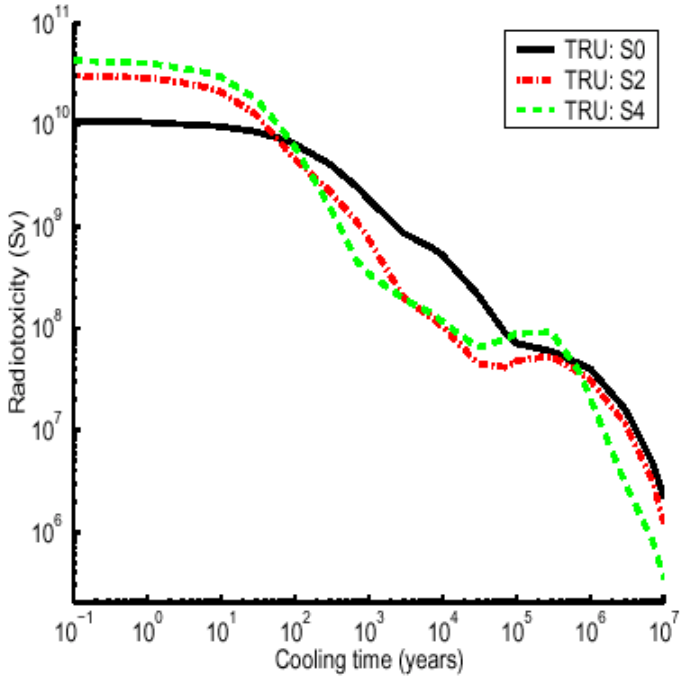
Again like in the Scenario S2, Scenario S4 shows that in order to obtain deeper burn-up of fissile materials the system should be run as long as possible even with decreasing core power. The initial reactor power cannot be kept constant due to large decrease in k_{eff} and also due to limitations on available accelerator power. Although, Scenario S2 ends up with slightly better burn-up rates both for ^{239}Pu and ^{240}Pu, at the very beginning it requires full separation of Np, Am and Cm isotopes. In the case of Scenario S4, the mass of ^{238}Pu is actually increased due to the presence of ^{237}Np in the fuel, what is not the case for Scenario S2. However, if the total mass of actinides is compared at the end of the fuel cycles, i.e. (Discharge S2 + 178.5 kg of Np, Am and Cm isotopes not irradiated) with (Discharge S4), the numbers are in favour of Scenario S4 by -57.5 kg.

The question which remains to be examined is the actual behaviour of fuel radiotoxicity for all different cases considered in this study. Figure 3 presents a change in radiotoxicity for Scenarios S0, S2 and S4 as described in the previous Section. It is important to note that the curve TRU: S2 contains the contribution to the radiotoxicity due to 178.5 kg of Np, Am and Cm isotopes (see Table 2) not irradiated in this particular scenario. In brief, there is no considerable gain in decreasing TRU radiotoxicity in a long term. As it is clearly seen from Figure 3, the total radiotoxicity of the irradiated fuel is higher by a factor of 3-4 during first 100 years of cooling. Later curves cross and irradiated fuel becomes less radiotoxic only by a factor of 4-5 after 1 000 years of cooling. After 100 000 years the radiotoxicity curves of irradiated fuel again approach the one corresponding to non irradiated TRU. Even more, Scenario S4 for a while gives even higher radiotoxicity compared to Scenario S0 as shown in the same Figure.

6. Conclusions and outlook

The problem of elimination of Pu isotopes has been addressed in terms of once-through fuel cycle. We confirm that the GT-MHR technology offers the potential to eliminate essentially all weapons-useful material present in nuclear waste. In addition, wide spectrum of plutonium isotopic compositions with or without isotopes of Np, Am and Cm prove GT-MHR potentials to use the plutonium as fuel without generating large amounts of minor actinides. In brief, we emphasise that significant levels of plutonium destruction (~99.5% for ^{239}Pu and ~84.4% for all Pu isotopes) can be achieved using critical and accelerator-driven subcritical thermal assemblies. The use of accelerator is essential to provide the needed neutrons for deep burn-up of ^{239}Pu and an important further destruction of ^{240}Pu and ^{241}Pu in the thermal regime without reprocessing.

Figure 3. **A change in radiotoxicity of TRU for different scenarios considered: TRU:S0 – no irradiation, TRU:S2 – Scenario S2 and TRU:S4 – Scenario S4. See Tables 2, 3 and 4 for detailed discharge compositions.**



On the other hand, none of the scenarios considered could reduce significantly the total waste radiotoxicity in the long term. Although only a small amount of higher actinides (mainly Am and Cm) is created during the fuel cycle, these elements are much more toxic than initial plutonium, what actually "compensates" the decreased total mass of actinides. Consequently, the question why other actinides but plutonium cannot be eliminated via fission in GT-MHR should be addressed. There are at least a few interlinked answers explaining this situation:

1. Np, Am and Cm are not efficiently fissioned in thermalised neutron flux simply due to very small σ_f/σ_c ratio.
2. Neutron fluxes available in GT-MHR in absolute value are rather low, say, of the order of $1\text{-}2\cdot10^{14}$ n/s/cm^2.
3. Typical fuel cycles of GT-MHR are rather short, i.e. 4-6 years.

Since other actinides with an exception of plutonium are more inclined to fission in a fast neutron energy spectrum, one could consider an additional fast stage, following a thermal stage, in order to eliminate the remaining actinides. Consequently, much bigger gain in reduction of the total radiotoxicity of the waste could be achieved. This is a subject of our future study, and the work along these lines is already in progress elsewhere [4,6,7].

REFERENCES

[1] International GT-MHR project, *Conceptual Design Report*, OKBM, Russia, report GT-MHR 100025 (1997).

[2] C. Rodriguez, J. Zgliczinski, and D. Pfremmer, *GT-MHR Operation and Control*, IAEA meeting on Gas Reactors, IAEA report (1994).

[3] A.J. Neylan, F.A. Silady, and A.M. Baxter, *Gas Turbine Modular Reactor (GT-MHR): A Multipurpose Passively Safe Next Generation Reactor*, General Atomics, LA-12625-M (1995).

[4] C. Rodriguez, A. Baxter, *Gas-cooled Transmutation of Nuclear Waste Using Thermal and Fast Neutron Energy Spectra*, Presentation at the Massachusetts Institute of Technology, USA (2000).

[5] X. Raepsaet, F. Damian, R. Lenain, and M. Lecomte, *Fuel Cycle Performances in High Temperature Reactor,* CEA and Framatome case study (1998).

[6] A.M. Baxter, R.K. Lane, and R. Sherman, *Combining a Gas Turbine Modular Helium Reactor and an Accelerator and near Total Destruction of Weapons Grade Plutonium*, AIP report, General Atomics, USA (1995) 347.

[7] C. Rodriguez *et al.*, *The Once-through Helium Cycle for Economical Transmutation in Secure, Clean and Safe Manner*, Paper 73 in the Proceedings of the 6th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, 11-13 December 2000, Madrid, Spain, OECD/NEA (Nuclear Energy Agency), Paris, France, 2001.

[8] L.S. Waters, *MCNPX™ USER's MANUAL*, Los Alamos National Laboratory, pre-print TPO-E83-G-UG-X-00001 (November 1999). See also: http://mcnpx.lanl.gov/.

[9] J. Briesmeister for Group X-6, *MCNP-A, A General Monte Carlo Code for Neutron and Photon Transport*, Version 4A, Los Alamos National Laboratory, pre-print LA-12625- M (1993).

[10] H.R. Trellue and D.I. Poston, *User's Manual, Version 2.0 for Monteburns, Version 5B*, Los Alamos National Laboratory, pre-print LA-UR-99-4999 (1999); H.R. Trellue, Private communication (April 2000).

[11] W.B. Wilson, T.R. England and K.A. Van Riper, *Status of CINDER'90 Codes and Data*, Los Alamos National Laboratory, pre-print LA-UR-99-361 (1999), submitted to Proc. of 4th Workshop on Simulating Accelerator Radiation Environments, September 13-16, 1998, Knoxville, Tennessee, USA; W.B. Wilson, private communication (May 2000).

[12] D. Ridikas, G. Fioni, P. Goberis, O. D'eruelle, M. Fadil, F. Marie, S. Röttger, *On the Fuel Cycle and Neutron Fluxes of the High Flux Reactor at ILL Grenoble*, CEA-report, DAPNIA/SPHN-00-52 (2000); available at http://www-dapnia.cea.fr/Doc/Publications/Sphn/sphn00.html.

[13] P. Goberis, *Modelling of Innovative Critical Reactors with Thermalized Neutron Fluxes in the Frame of the Mini-Inca Project*, rapport du stage at CEA Saclay, DAPNIA/SPHN-00-68 (2000); available at the request by e-mail: D.Ridikas (ridikas@cea.fr).

[14] G. Fioni, O. D'eruelle, M. Fadil, Ph. Leconte, F. Marie, D. Ridikas, *Experimental Studies of the Transmutaion of Actinides in High Intensity Neutron Fluxes*, Proc. of the 10th International Conference on Emerging Nuclear Energy Systems, ICENES 2000, Petten, the Netherlands, September 25-28, 2000, p.374.