Experimental Studies of the Transmutation of Actinides in High Intensity Neutron Fluxes

G. Fioni, O. Dérieuille, M. Fadil, Ph. Leconte, F. Marie, D. Ridikas
Commissariat à l’Énergie Atomique
CEA/Saclay, DSM/DAPNIA/SPhN
Gif-sur-Yvette (France)
gfioni@cea.fr ; oderuelle@cea.fr ; mfadil@cea.fr ; fmarie@cea.fr ; dridikas@cea.fr

Keywords: Transmutation, Cross sections, Thermal Neutrons, High-Intensity Fluxes, Integral Measurements, Helium Reactor, Molten Salt

Abstract

High intensity neutron fluxes are of major interest for nuclear waste transmutation. Innovative fuel cycle reactors, operated in critical or sub-critical modes, provide promising alternatives for a reduction of the long term radiotoxicity and of the proliferation risk associated with nuclear waste. Due to the significant uncertainties in the nuclear data libraries, integral experiments are essential for a correct understanding of the transmutation process and for a precise estimate of the neutronic parameters of innovative systems.

1. Introduction

Actinides can be transmuted by neutron induced nuclear fission into short-lived fission fragments with a gain in long term radiotoxicity of spent fuel. In principle any neutron source can be used for nuclear waste transmutation. Critical nuclear reactors are the most natural candidate, even if accelerator driven systems (ADS) present significant advantages both for the transmutation capacity and for the system safety when using minor actinides as fuel (Fioni et al., 2000).

The main parameter in transmutation is the neutron energy spectrum of the system, and most of the proposed designs reflect the need for a variety of different spectra in order to have a more efficient and economically viable transmutation. Unluckily, a unique solution is impossible because the capture and neutron cross sections vary considerably from one isotope to another. If fast neutron spectra present the advantage of higher neutron excess, thermalised spectra have the merit of higher cross sections. This results in more compact core configurations with faster burn-up of the fissile material. For a given thermal power of the system, the in-core mass can be up to a factor of 20 lower in thermalised systems (Lelievre 1998). The lower neutron excess requires the presence of highly fissile material, as $^{239}$Pu or $^{235}$U, or on-line recycling of the fuel in order to eliminate fission products which depress the reactivity of the system.

Due to the number of rare isotopes playing a major role in the innovative fuel cycle, and to the impossibility to carry out comprehensive differential cross section measurements, integral experiments are necessary to assess the quality of computer code simulations that use the existing nuclear data libraries. Most of the data for minor actinides contained in the main nuclear data libraries are based on pure theoretical evaluations, due the lack of experimental data. Even when experimental data exist, it is rather difficult to assess the quality of measurements carried out more than 30 years ago, mostly for military applications and for which very little information is available. The presence of a number of isotopes with long-lived metastable states, often with longer half-lifes
than the corresponding ground state, get the interpretation of old experiments even more puzzling. An extreme example is provided by the $^{242}$Am thermal neutron capture cross section, where a factor of 20 difference exist between the American (ENDF-B/BVI) and the European (JEF 2.2) nuclear data libraries. This isotope plays a dominant role in the neutronics of high intensity thermalised systems and only a recent integral experiment at ILL in Grenoble was able to solve this outstanding problem (Fioni et al., 1999, Aubert et al., 2000). The computed transmutation efficiencies of different systems can be strongly affect by the quality of nuclear data, and may result in wrong conclusions.

2. Innovative Systems with Thermalised Neutron Spectra

Several innovative fuel cycle systems are based on thermalised neutron spectra, as the GT-MHR reactor, the AMSTER (Lecarpentier et. al., 1999) and the TIER (Bowman, 1999) molten salt reactors. They all present interesting features and we are carrying out detailed simulations in the frame of the Mini-Inca project (Goberis, 2000). The interesting point is that they can all work as critical reactor as well as accelerator driven sub-critical systems.

Two computing code sequences are used, both based on the MCNP code (Briesmeister, 1993) for the calculation of the neutron transport. As it will be shown in the following section, the evolution of the fuel composition must be taken into account to assess correctly the neutronics of the system. The first method is based on the coupling between MCNP and the CINDER code (Wilson et al., 1999), while the second make use of the recent Monteburns code (Trellue et al., 1999). This last code automatically couples MCNP to the evolution code ORIGEN 2.1. A number of computational tests showed the consistency of the results obtained with the two approaches (Ridikas et al., 2000).

In the following, only the results obtained for the GT-MHR reactor will be presented, as the simulations for the other systems are not completed.

2.1 Gas Turbine Modular Helium Reactor (GT-MHR)

The GT-MHR is based on an innovative 600 MWth graphite moderated reactor (Neylan et al.,1995, International, 1997). The cooling is assured by high-pressure helium, whose outlet temperature is 850 degrees Celsius. This allows the coupling with a high efficiency gas turbine, providing an overall energy conversion efficiency between 45 and 47 %, face to about 33% of conventional light water reactors.

The GT-MHR was developed in a joint project by USA, France, Japan and Russia in order to reduce the amount of weapons grade and civil Plutonium (WGPu and CPu respectively). Beside these Plutonium based fuel cycles, it can make use of Uranium based fuels in the form of $^{235}$U/$^{238}$U and $^{235}$U/Th (U5/U and U5/T respectively).

One of the most interesting feature of this reactor is constituted by the fuel which is in a form of graphite coated micro-particles: a spherical kernel of a fissile/fertile material is coated by pyrocarbon and silicon carbide layers (TRISO coating), for a total diameter of about 300 µm. The high resistance to the temperature and to radiation damage allows burn-up rates higher than 90%, without a significant release of fission fragments.

The modularity of the reactor, which is formed by a repetition of a limited number of identical modules, allows the integration of a spallation target module for the operation in sub-critical mode. In the following we will describe the results obtained with the Monteburns code for the operation of the GT-MHR in critical mode for WGPu, CPu and U5/U (with a 14% enrichment) fuels. The simplified model of the reactor used for the simulations is shown in Fig. 1. It consists of the reactor core (C), the inner reflector (Ri), the side reflector (Rs), the top and the bottom reflectors (Rt and
Rb respectively) and the reactor vessel (V). The inner and the outer core diameters are respectively 2.96 and 4.84 m, and the height of the fuel part is 8.0 m, for a total core volume of 92.1 m$^3$. The total height of the core is 10.9 m, including the top and bottom graphite reflectors. The reactor vessel consists of stainless steel 20 cm thick, with an outer diameter of 7.5 m and a height of 20.22 m.

In Fig. 2 the evolution of the $k_{\text{eff}}$ is plotted as a function of the number of full-power days (fpd) for the three fuels mentioned above. It can be observed how strongly the length of the reactor cycle depends on the type of fuel. It should be stressed that no partial refuelling of the core takes place in a GT-MHR. In fact, to assure a negative temperature coefficient of the reactor, a variable amount of natural erbium (Er$_2$O$_3$) is present in the core for the three types of fuel. Due to the high neutron capture cross section of $^{167}$Er, it acts as a burnable poison to control the excess of reactivity during the reactor operation. The control rods, whose movement was not taken into account in the simulation, counteract the excess of reactivity shown in Fig. 2.

Figure 1: Axial view of the GT-MHR simplified model as used for the numerical simulations.
Figure 2: Evolution of $k_{\text{eff}}$ as a function of time in the GT-MHR for three different fuel types. The initial and final composition for the WGPu, CPu and U5/U fuels is given in Table 1. It can be observed the remarkable $^{239}$Pu reduction higher than 95% and, for the U5/U fuel, a reduction about 69% is obtained for the $^{235}$U. A even deeper burning of the fuel can be obtained by operating the reactor in sub-critical mode for another year.

Table 1: Initial and final composition for the WGPu, CPu and U5/U fuels

<table>
<thead>
<tr>
<th>Isotope</th>
<th>WGPu Fuel</th>
<th>CPu Fuel</th>
<th>U5/U Fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Load [kg]</td>
<td>Discharge [kg]</td>
<td>Load [kg]</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>630</td>
<td>197</td>
<td>696</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>3870</td>
<td>3660</td>
<td></td>
</tr>
<tr>
<td>$^{236}$U</td>
<td>0</td>
<td>73.8</td>
<td></td>
</tr>
<tr>
<td>$^{237}$Np</td>
<td>0</td>
<td>3.33</td>
<td></td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>11.9</td>
<td>20.4</td>
<td>0</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>658</td>
<td>13.7</td>
<td>690</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
<td>37.5</td>
<td>36.3</td>
<td>274</td>
</tr>
<tr>
<td>$^{241}$Pu</td>
<td>54.1</td>
<td>156</td>
<td>96.8</td>
</tr>
<tr>
<td>$^{242}$Pu</td>
<td>46.3</td>
<td>156</td>
<td>96.8</td>
</tr>
<tr>
<td>$^{243}$Am</td>
<td>6.4</td>
<td>33.2</td>
<td></td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>0</td>
<td>28.4</td>
<td></td>
</tr>
<tr>
<td>TOTAL Actinides</td>
<td>696</td>
<td>165</td>
<td>1190</td>
</tr>
</tbody>
</table>
The evolution as a function of the time of the CPu fuel composition is given in Fig. 3. During the 4 years cycle of the reactor, the strong reduction of $^{239}\text{Pu}$ and $^{241}\text{Pu}$ is accompanied by an increase of $^{242}\text{Pu}$ and $^{244}\text{Cm}$.

![Figure 3: Evolution of the fuel composition due to the burn-up in the GT-MHR reactor, loaded with civil Plutonium. It should be noted that after 4 years the reactor is shutdown and the isotopic evolution is only due to nuclear decay.](image)

The neutron energy spectra averaged over the core region for the CPu fuel are shown in Fig. 4 for four different burn-up rates, after 180, 540, 900 and 1260 full power days. The thermal component of the spectrum change from 10 to 31%, due to the burn-up of the burnable poison (Er) and, to a lesser extent, to the modification of the fuel composition.
Figure 4: The GT-MHR neutron energy spectrum for 4 different burn-up of the CPu fuel. The integrated neutron fluxes in the core are of the order of $1-2 \times 10^{14} \text{ n/s/cm}^2$

Figure 5: One group neutron induced fission cross sections for the GT-MHR as a function of time. The CPu fuel was used in these simulations

These spectral modifications can strongly affect the spectrum averaged neutron cross sections. The one-group fission cross section obtained from MCNP as a function of the reactor operating days are
plotted in Fig. 5 where an increase up to a factor of four is observed. The burn-up simulations were carried out taking into account these variations as a function of time. Simulations carried with constant cross section values would lead to completely wrong results. This aspect must not be neglected, especially for innovative systems that present variations in the core composition which largely exceed the ones of conventional light water reactors.

A precise knowledge of the neutron cross sections at the thermal and epithermal energies is essential, as several of the innovative systems are based on moderated neutron induced reactions.

3 The Mini-Inca Project

The quest for integral measurements relevant for transmutation systems was the main trigger for the Mini-Inca project (Fioni, 1998) at Directorate of Science of Matter of the French Atomic Energy Authority (CEA/DSM). It aims to develop a set of experimental methods and computational procedures to carry out integral measurements to assess the transmutation potential and the average nuclear parameters of specific isotopes in given neutron spectra. This will be achieved by measuring the isotopic composition of the sample before and after irradiation in a measured neutron flux.

To obtain valuable data, we have therefore to carry out a new type of integral experiments based on:

- small mass samples of about 10 µg, to avoid perturbations of the local neutron flux;
- the use of high intensity fluxes, to have access to very short lived isotopes by multiple neutron capture;
- the possibility to carry out nuclear spectroscopy measurements shortly after the irradiation, when short-lived isotopes are still present;
- the use of neutron spectrum monitors irradiated together with the sample, and the use of on-line neutron detectors to follow the variation of the flux intensity as a function of time.
Long irradiation of mono-isotopic or a known mixture of transuranic elements and fission fragments will provide the transmutation rates in given high intensity neutron spectra. While this type of measurements will not be very useful to determine nuclear parameters, they will provide the effective transmutation efficiencies and the equilibrium compositions. On the opposite, short irradiation of high-purity mono-isotopic samples will allow a precise determination of nuclear parameters as neutron cross sections, branching ratios and half-lives.

The isotopic composition will be obtained by a number of complementary techniques, from classical off-line mass spectrometry (TIMS and ICP-MS) to alpha-gamma nuclear spectroscopy. A schematic diagram of the project is given in Fig. 6, where most of the techniques that are used and that will be developed are shown. A particular effort has been devoted to the development of a new type of fission μ-chambers (Fadil, 2000) that will permit to follow on-line the fission rate of a sample during a 50 day irradiation. A precise experimental information on the reactivity of the sample as a function of the evolution of the isotopic composition will thus be obtained.

4 Experimental Set-up

The choice of the ILL high-flux reactor as the first experimental facility for the Mini-Inca project is mainly due to the unique possibility to dispose of several different neutron spectra, obtained by changing the distance between the sample and the fuel element. Additionally it provides the highest thermal flux in the world, which makes possible high accuracy measurements in the thermal energy region and to carry out experiments at the same intensities foreseen for a transmutation system which can approach $3 \times 10^{15}$ n/s/cm².

4.1 Variable Flux Facility

The energy distribution of neutrons generated by fission of $^{235}$U can be parameterised as an evaporation spectrum with an average energy of about 2 MeV. Inside the reflector tank of the ILL reactor, neutrons are moderated by the heavy water, where the scattering length is about 20 cm.
Different neutron spectra will be present at different distances from the fuel element, with an increase of the moderation as a function of the distance. Starting from about 60 cm from the fuel element edge, the spectrum is essentially a Maxwellian distribution at the moderator temperature. In Fig. 7, the neutron densities obtained at different distances from the axes of the reactor are given as a function of the neutron energy. The spectra at about 25 cm, present similarities, both in intensity and in shape, to the one obtained for moderated transmutation systems, as shown previously.

A schematic view of the V4 inclined beam tube, which has been used for the MCNP code simulations, is given in Fig. 8.
The samples will be positioned at four different distances along the channel, to have access to three epithermal fluxes and to one thermal flux. At the closest position the total flux will be about $2 \times 10^{15}$ n/s/cm$^2$, half of which is thermal. The pure thermal beam position will have a flux of about $5.5 \times 10^{14}$ n/s/cm$^2$.

4.2 Thermal Flux Position

An essential complement to the variable flux facility is provided by the H9 beam tube, which gives access to a neutron flux of about $5 \times 10^{14}$ n/s/cm$^2$ with a thermalization coefficient higher than 98%. It is already equipped with a source changing facility to get at a distance of about 60 cm from the edge of the fuel element. The beam tube is connected to the Lohengrin parabola mass spectrometer for on-line measurements of unslowed fission fragments (Moll, 1975). The evolution of the composition of a minor actinides sample can be determined on-line by measuring the alpha particles emitted by decay of isotopes formed during the irradiation. The variation of the fission rate will be measured at reference isobaric line and it will permit the determination of the fission cross section. At the same time, the measurement of the energy distribution of the fission fragments will provide information on the chemical and physical behaviour of the target material. The presence of sputtering of the target material, or a sudden lost of material due to the extreme thermal conditions during the irradiation, can be detected.

To perform off-line nuclear spectroscopy of irradiated samples, the existing target changer has been modified to host a new vacuum chamber (Deruelle et al., 2000) that allow to position the sample in front of a detection system, with an absolute precision of 0.1 mm. Two detectors are used for gamma-rays and alpha particles. The gamma detector is a high resolution coaxial germanium with a relative efficiency of 15%. A 0.5mm thick carbon-epoxy end-cup allows to have a wide energy range of gamma detection from
about 20 keV to several MeV. It has a resolution of 0.825 keV (FWHM) at 122 keV and 1.8 keV at 1.33 MeV with a peak/Compton ration of 46:1. The detector and its dewar can move from 30 to 70 cm and in the fraction of solid angle, \((\Omega/4\pi)\), will vary from 2.10^{-3} to 3.10^{-4}. The alpha detector is a high resolution PIPS (Passivated Implanted Planar Silicon) of active area of 100 mm² depleted on 300 µm. The resolution is 11.8 keV for 5.486 MeV alpha particles. It can move from 0.4 cm ((\Omega/4\pi) = 0.1 ) to 30 cm ((\Omega/4\pi) = 10^{-5}) from the irradiated sample.

Due to the high counting rate (>50 kHz) foreseen in a number of measurements, a dedicated high speed electronics is used. A Digital Signal Processor handles all signals after the pre-amplification stage. A transistor-reset preamplifier is used to suppress pile-up effects on the resolution of the Germanium detector. Preliminary tests show a constant resolution up to 50 kHz and a gradual degeneration up to a factor of three at 100 kHz.

Both detectors can be moved to choose the most suitable solid angles and consequently the counting rates. The chamber and the detectors are shielded by a 5 cm thick lead wall and by a combination of borated polyethylene and boron carbonate to reduce the gamma and neutron backgrounds. The complete spectroscopic system in under commissioning and the first experiments are scheduled in November 2000.

A number of measurements will be carried out at this thermal energy position, with an accuracy in the 5-15% range (Marie, 2000). The samples which are at present available include \(^{239,241,242}\text{Pu}, ^{237}\text{Np}, ^{241,243}\text{Am}, ^{244,245}\text{Cm} and ^{249}\text{Cf}, and the experimental programme will last about 24 months.

5. Conclusions

Integral measurements of transmutation efficiencies are essential to assess the performances of transmutation and innovative fuel cycle systems operated with high intensity thermalised neutron fluxes. The Mini-Inca installation will provide, in the near future, unique experimental data for a basic understanding of the physics of transmutation. High quality cross section values will be measured, increasing both the quality of existing nuclear data libraries and the consistency of simulations.

References

Briesmeister J. for group X-6, Los Alamos National Laboratory, LA-12625-M (1999)
Deruelle O. et al., Proceedings of the 8th ISINN Conference, Dubna (Russia), (2000) in press
Lecarpentier D., Vergnes J., Tetard Ph., EDF-France, reports HT-10/99/020A, HT-10/99/022A

Moll E. et al., Nucl. Instr. and Meth. 123 (1975) 615


