TRANSMUTATION STUDIES OF MINOR ACTINIDES IN HIGH INTENSITY NEUTRON FLUXES

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Abstract

Integral measurements of nuclear data and of the transmutation potential in specific neutron fluxes, constitute the fastest and essential way to overcome to the large uncertainties present in the nuclear data libraries. In the frame of the activities of the Directorate for Science of Matter (DSM) of the French Atomic Energy Authority (CEA), we propose a new project to carry out integral measurements relevant for nuclear waste transmutation systems.

A new beam tube will be installed to irradiate actinides and fission fragment samples at different distances from the fuel element of the ILL reactor. Variable neutron energy spectra could then be obtained by choosing the distance between the sample and the fuel element, opening the way to the determination of the ideal physical conditions to incinerate nuclear waste in hybrid transmutation systems.

Introduction

Social acceptability of nuclear energy is strongly related to the problem of long-lived nuclear waste produced during the burn-up of the nuclear fuel. By successive neutron captures, transuranic elements are formed from the initial uranium and plutonium isotopes, while by nuclear fission a large variety of light and medium mass isotopes are produced.

Several solutions aiming at the reduction of the long-term radiotoxicity by the transmutation of long-lived isotopes into stable or short living elements have been studied in the last years. A general consensus indicates that only neutron-induced reactions are adequate, due to the combination of high cross sections and to their lower energetic cost of production. Other options that were once suggested, as direct spallation by high-energy protons and gamma-ray induced reactions, seem to be economically non-viable, since the destruction of waste would require more energy than produced when generating this waste.

In principle any neutron source can be used for nuclear waste transmutation. Actinides can be transmuted by nuclear fission mostly into short living nuclei with a considerable gain in long term radiotoxicity. Long-lived fission products can be transmuted into stable isotopes by direct or successive neutron captures followed by beta decays. A clear example is constituted by the reaction.

$$^{99}Tc(n,\gamma)^{100}Tc \xrightarrow{T_{1/2}=15.8s} {}^{100}Ru$$

where the long-lived ($T_{1/2}$ =2.1 x10^5 years) ^{99}Tc is transmuted in 15.8 seconds into the stable ^{100}Ru.

Fission reactors constitute the most direct source of neutrons, and studies on the transmutation capabilities of thermal or fast reactors are being conducted in several countries. While the incineration of military plutonium seems to be possible in ordinary and fast reactors, nuclear waste transmutation is more difficult. In fact, ordinary PWRs do not have suitable neutron excess to transmute nuclear waste, unless the uranium enrichment is increased significantly. Fast reactors have a more favourable neutron balance, but safety considerations limit the amount of plutonium isotopes and minor actinide fuel that could be irradiated.

The neutron cross-sections for capture and fission reactions decrease significantly as a function of the neutron energy. Hence, a thermal neutron flux will provide higher transmutation rates for a given mass than epithermal and fast spectra. Nevertheless, as the transmutation of minor actinides is achieved by nuclear fission and not all isotopes are fissile at thermal energy, an essential parameter to be evaluated is the ratio between fission and capture cross-sections, which is larger in a fast neutron flux. Therefore fast spectra are preferable in order to avoid the generation of high Z actinides and to obtain higher neutron excess. The price to pay is a larger fuel inventory in the installation for a given thermal power.

Without going further into the details of this problem, it should be stressed that the choice of the most suitable neutron spectrum depends on the characteristics of each specific isotope and that systems where thermal and fast spectra coexist in the same installation are under study. To date no decision can be based on experimental bases, but only on computer simulations of the transmutation process.

The major problem in a clear assessment of the "ideal" neutron flux is constituted by the very poor knowledge of nuclear cross section data for most of the minor actinides and fission fragments concerned. Though a general agreement exists in the most widely used nuclear data library (JEF-2.2, ENDF-B/VI and JENDL 3.2) for nuclei that play a major role in the "conventional" fuel cycle, the situation concerning rarer isotopes like Am, Cm and Np is totally unsatisfactory. Large discrepancies exists in both thermal and fast energy regions, originating in large differences on the neutronics characteristics of the systems.

An extreme example is given by the ^{242gs}Am cross section where a difference of a factor of 20 existed between ENDF and JEF, making impossible any prediction on the possibility of transmutation of ^{241}Am in thermal fluxes. A recent experiment at ILL has been able to fix this large discrepancy [1].

An essential contribution to the solution of this scientific debate is constituted by integral measurements, where the evolution of a sample is measured for a specific neutron spectrum. In this frame the Mini-Inca project constitutes a unique experimental approach to carry out experiments in conditions which are representative of a thermal neutron based incineration systems.

The mini-Inca project

The Mini-Inca project aims to develop a set of experimental methods and computational procedures to carry out integral measurements to assess in a fast and reliable way 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 after irradiation in a measured neutron flux. The evolution of a sample in a neutron flux is given by a linear system of differential equations [2]:

$$\frac{dN}{dt} = -MN \quad (1)$$

where : M is a square matrix where the non-diagonal elements give the reaction rates j->i and N is a vector giving the isotope population N_i for a given isotope i. The reaction rates Mij are given by the neutron induced reaction [$\sigma\,\phi$] or by the decay constants [$\lambda = \ln 2 / T_{1/2}$].

If the concentrations N_i and the neutron flux are known, a number of nuclear parameters can be obtained by a simple fit between the experimental data and the solutions obtained by solving the system (1).

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

- Small mass samples, to avoid any problem related to corrections of the local flux due to the sample influence.
- A set of experimental methods from nuclear spectroscopy to mass spectrometry, to obtain in the fastest way the sample composition.
- The use of high intensity fluxes, to have access by multiple neutron capture to very short-lived isotopes.
- 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 detectors to follow the variation of the flux intensity as a function of time.

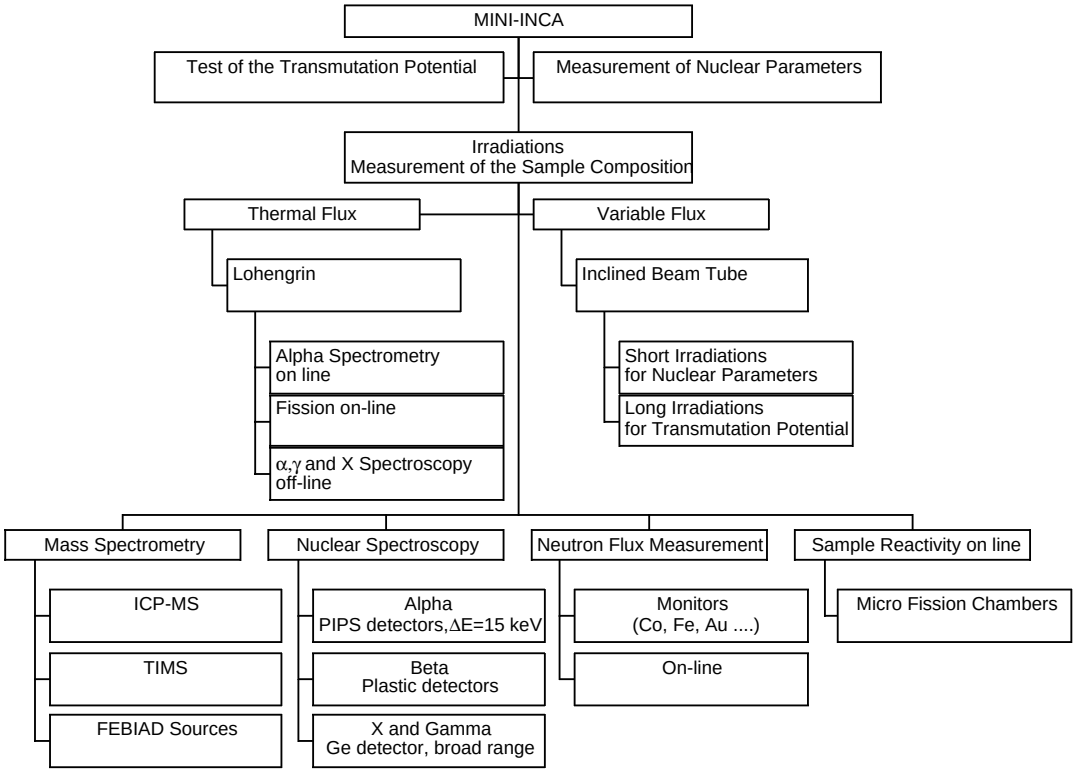
Two type of measurements are foreseen:

1. Long irradiations of mono-isotopic or a known mixture of transuranic elements and fission fragments, to determine the transmutation rates in given high intensity neutron spectra. These type of measurements will not be very useful to determine nuclear parameters, but they will indicate the effective transmutation possibilities and the equilibrium compositions.
2. Short irradiations of high-purity mono-isotopic samples, for 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 new quasi-on-line mass spectrometry with FEBIAD sources [3], and from alpha-(beta)-gamma nuclear spectroscopy [4].

A schematic view of the project is given in Figure 1, where most of the techniques that will be used and developed are indicated.

Figure 1. **Schematic view of the Mini-Inca project.**



Dedicated irradiation facilities at ILL

The choice of the ILL reactor as the first facility to start 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

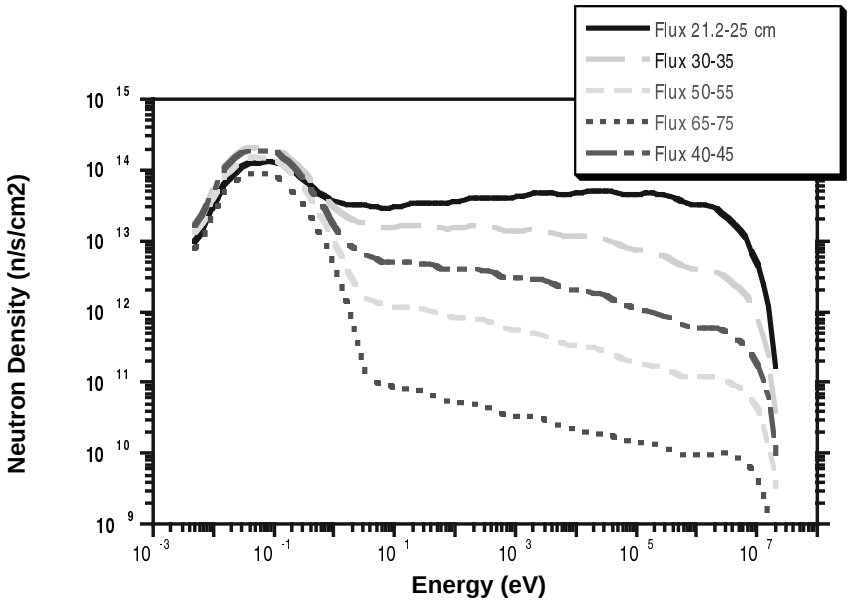
the world, which makes possible high accuracy measurements in the thermal energy region and to carry out experiments in at the same intensities foreseen for a transmutation system which can approach $3*10^{15}$n/s/cm^2.

Variable flux facility

The energy distribution of neutrons generated by fission of ^{235}U can be parametrized 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. The moderator partly reflects the thermalized neutrons towards the fuel element. It is evident that 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 Figure 2, the neutron densities obtained at different distances from the axes of the reactor are given as a function of the neutron energy.

Figure 2. **Neutron Spectra at ILL for different distances from central axes of the reactor. The outer edge of the fuel element is located at 20.68 cm from the axes of the reactor.**



The plotted values correspond to an average flux $\langle\phi\rangle_i$ for a given energy interval $(E_i - E_{i-1})$ defined as:

$$\langle\phi\rangle_i = \int_{E_{i-1}}^{E_i} \phi(E)dE$$

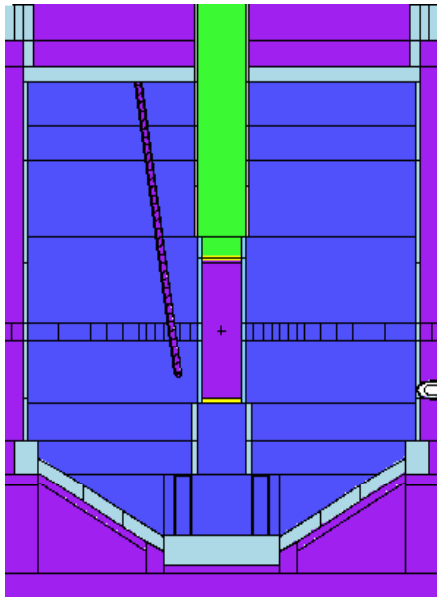
with an energy bin such that:

$$\frac{\Delta E}{E} = cst$$

The spectra at about 30 cm, correspond both in intensity and in shape, to the one obtained for a strongly thermalized transmutation system [5]. The interest in making measurements in these kinds of thermal neutron fluxes is therefore evident. Additionally by measuring integral nuclear parameters with different spectra, we can have access to the average energy dependence of neutron cross sections.

A scheme of the installation of an inclined beam tube, which has been used for the MCNP code simulations, is given in Figure 3.

Figure 3. **Scheme of the installation of an inclined beam tube, which has been used for the MCNP code simulations**



The goal is to install a very small channel of about 5 cm outer diameter, in order to minimise the anti-reactivity. Preliminary calculations indicate a loss of about 300 pcm for an Ag3Net (Al alloy) getting at 5 cm from the edge of the fuel element and filled with heavy water. This will correspond to a loss of about 12 hours of reactor cycle.

The samples have to 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 3×10^{15}n/s/cm^2, half of which is thermal. The pure thermal beam position will have a flux of about 5×10^{14}n/s/cm^2.

In order to minimise all manipulations of the irradiated sample, automatic irradiation systems are under study. The sample will be brought at the chosen irradiation position either by mechanical or by hydraulic systems. After irradiation it will go directly inside a dedicated hot-cell for conditioning either before shipping it to a radiochemistry laboratory or before being introduced automatically in a nuclear spectroscopy set-up.

Thermal flux position on H9

A valuable complement to the variable flux facility, is provided by the H9 beam tube which gives access to a neutron flux of about 5×10^{14}n/s/cm^2 with a thermalization coefficient higher than 98%. It is already equipped with a source changing facility to get 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 unslowed fission fragments.

We also plan to use the H9 beam tube both for on-line measurements with Lohengrin and for off-line nuclear spectroscopy using a modified target changer.

The evolution of the composition of a minor actinide sample can be followed on-line by looking at the alpha particles emitted by decay of the isotopes formed during the irradiation. The formation and the decay rates of several isotopes could then be followed. The global fit of the obtained intensities will enable a comparison with the predictions obtained by solving the system (1) describing the sample evolution from different nuclear data libraries.

Another advantage of using Lohengrin lies in the possibility of also following the fission rate by looking at a given isotopic chain. In this case the target gives the main problem: beside the optimisation of the thickness and the geometry for the alpha and the fission spectrometry, the sputtering of the target material can constitute a major limitation. A new type of isotope deposition on titanium or graphite supports must be envisaged.

Another possibility of the H9 beam tube, is to use the target changer just to irradiate a sample and recover it to perform off-line nuclear spectroscopy. This requires a modification of the source changer and the installation of a "reaction chamber" on the changer tube. The chamber will be used under primary vacuum and it will house detectors to perform alpha-gamma spectroscopy. The main advantage of an installation of this type is that it will not be necessary to perform complicated manipulations of the sample and that the irradiation can continue after an intermediate spectroscopic measurement. Additionally, after the measurement the sample can be put into the waste container of Lohengrin.

A number of measurements are possible at this thermal energy position, with accuracies higher than 5 per cent.

Conclusions

The realisation of the proposed project will contribute significantly to the development of nuclear waste transmutation systems. It will provide the scientific and technical communities with a set of experimental procedures to assess in a fast and reliable way the transmutation potential and nuclear data for specific neutron fluxes.

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