Development of Fission Micro-Chambers for Nuclear Waste Incineration Studies

M. Fadil a, Ch. Blandin b, S. Christophe c, O. Déruelle a, G. Fioni a, F. Marie a, C. Mounier d, D. Ridikas a, JP. Trapp b

a DSM/DAPNIA/SPhN – CEA/Saclay, 91191 Gif-sur-Yvette, France
b DRN/DER/SSAE/LSMN, CEA/Cadarache, 13108 St. Paul-lez-Durance, France
c Ecole Nationale Supérieure de Physique, 38026 Grenoble, France
d DRN/DMT/SERMA, CEA/Saclay, 91191 Gif-sur-Yvette, France

Abstract. The INCA (INCineration by Accelerator) project of the Directorate for Science of Matter of the French Atomic Energy Authority (CEA/DSM) aims to outline the ideal physical conditions to transmute minor actinides in a high intensity neutron flux obtained either by hybrid systems or innovative critical reactors. To measure on-line the incineration rates of minor actinides, we are developing an innovative double deposit fission chamber (DDFC) working in current mode. Our method is based on a comparison between the isotope under study and a reference material whose nuclear parameters are well known, as 235U and 239Pu. This new fission chamber will be used in the High Flux Reactor in Grenoble/France in a neutron flux of 1.2 $10^{15}$ n.cm$^{-2}$.s$^{-1}$ for 50 days, the operating cycle of the reactor. These specific experimental conditions require substantial modifications of the existing chambers. The first experiment will be carried out in fall 2000.

Keywords: Transmutation; Fission chamber; Ionisation gas; Incineration

1. Introduction

The incineration of transuranic elements by neutron induced fission is a very promising way to reduce long-term radiotoxicity of nuclear waste. The Mini-Inca aims to outline the ideal physical conditions to transmute minor actinides, mainly $^{241-243}$Am, $^{237}$Np and $^{244-245}$Cm [1]. For some actinides there are large discrepancies of neutron cross sections taken from different evaluated nuclear data libraries. These cross sections will play a dominant role in transmutation systems. For instance, a factor 20 was pointed out for the $^{242}$Am thermal neutron capture cross section [2] from JEF-2.2 (5500 barns) and ENDF-B/VI (250 barns) libraries. Computer simulations can lead to controversial results depending on the nuclear data library that was used. To access experimentally the incineration rate of minor actinides, and to provide an unambiguous experimental reference, Fission Micro-Chambers are of great interest.

2. Use of Fission Micro-Chambers for Mini-Inca

In order to measure on-line the fission rate of actinide targets, we are developing innovative Double Deposit Fission Chamber (DDFC). Up to 50 days of irradiations will be carried out in the V4 inclined beam tube of the High Flux Reactor (HFR) at the Laue-Langevin Institute (ILL). This tube will permit us to irradiate samples in a high intensity variable neutron spectra. The thermalisation of the spectrum varies from 80 to 98 % depending on the position along the tube, while the total intensity changes correspondly from 1.8 to 0.6 $10^{15}$ n.cm$^{-2}$.s$^{-1}$. We estimate that a good sensitivity to the fission cross sections can be obtained by measuring on-line the fission rate of a given sample with respect to a reference isotope: $^{235}$U or $^{239}$Pu. In such a way the incineration rate of the sample under study can be followed on-line, providing valuable information including the evolution of its isotopic composition. This experimental technique can also be used to measure the incineration rate of a given isotopic mixture, and dedicated DDFC using recycled MOX fuel are under consideration.

To realise these measurements, micro-fission chambers are the best possible choice. These detectors have the advantage to give a response that is directly proportional, in saturation mode, to the fission rate. However, up to now, classic fission micro-chambers were used in neutron fluxes whose intensities rarely exceed $10^3$ n.cm$^{-2}$.s$^{-1}$. Our experimental conditions require the development of a new generation of fission micro-chambers, which can operate in a flux of some $10^5$ n.cm$^{-2}$.s$^{-1}$. In this high
flux, to avoid the pulse pile-up, the Chamber must be designed to operate in current mode. We are developing a Simple and a Double Deposit Fission Chambers which can fulfil to this requirement.

At a first stage, we will use a set of four Simple Deposit Fission Chamber (SDFC), with $^{235}$U as an active deposit, but with different gases as described below. The neutron flux intensity will be measured in the V4 channel with a period of a few hours for four days. That will enable us to define the best operational conditions of the chamber.

After the qualification and the measurement of the flux intensity by SDFC, the new DDFC will be used to measure the fission rates in comparison with a reference material, according to the following expression:

$$\frac{R_X}{R_{ref}} = \frac{I_X}{I_{ref}},$$

where

$R_X$ : fission rate of the actinide X (fissions.cm$^{-3}$.s$^{-1}$),

$R_{ref}$ : fission rate of the reference element (fissions.cm$^{-3}$.s$^{-1}$),

$I_X$ : saturation current due to the actinide X (µA),

$I_{ref}$ : saturation current due to the reference element (µA).

### 3. Operating conditions

The operating state of a fission chamber, i.e. the saturation current and the operating voltage, is determined by four factors: the gap, namely the difference between the cathode and the anode radii, the mass of the deposit, the nature and the pressure of the gas filling. We have to find a compromise between these factors to define the best operating conditions of the fission chamber in the neutron spectrum of the HFR.

#### 3.1 Electric field distortion

Ideally the electric field in the cylindrical chamber must be radial. The operation characteristic of the chamber is strongly related to the distortion of this field. This is mainly due to space charge effects, which induce a distortion $\delta E$ that can be written as:

$$\delta E(r, N_0, C) = \left[N_0 \frac{e}{4\varepsilon_0} \left(\frac{r^2 - 2r_a^2}{\mu_+} + \frac{r^2 - 2r_c^2}{\mu_-}\right) + \frac{C}{r^2}\right]^{1/2},$$

where $r_a$ and $r_c$ are respectively the anode and cathode radii;

$\mu_+$ et $\mu_-$ the mobilities of the charges depending on the nature of the gas;

$N_0$ the density of ions-pairs which depends on the pressure and the type of the gas;

$C$ is a constant depending on $N_0$ and on the gap.

All these factors have an influence on the operating state of the chamber and have to be optimised for the present application.

#### 3.1 Influence of the geometry and of the filling gas

Let $\Phi_s$ be the minimum flux at which there is no saturation current. In Fig. 1 we present the variation of $\Phi_s$ as a function of the gap, for a chamber with a cathode radius of 1.75 mm and for different masses of the $^{235}$U active deposit. We can observe that the greater the gap, the smaller $\Phi_s$ is, since the distortion $\delta E$ is proportional to $1/r^2$.

We can also observe that the deposited mass has a very similar effect, as an increase in the mass results in a higher number of fission fragments in the chamber and consequently to a higher space charge effect.
The nature of the filling gas plays an essential role on the saturation flux, as shown in Fig. 2, where $\Phi_s$ is given for 7 different gases. A mass of 20 $\mu$g of $^{235}$U and a gap of 0.5 mm were taken for these calculations. The differences in the results are mainly related to the ionisation potential and to the mobility of the ions in the gas.

We can first observe the Ramsauer effect [3] in the case of the two gas mixtures used. The addition of even a small percentage of other gases to the argon or to the neon increases the mobility of the charges in the gas, leading to a decrease of the space charge. This is clearly seen in Fig. 2 comparing Ar with Ar+0.5% C$_2$H$_2$ and Ne with Ne+0.5% Ar.

The other way to reduce the number of ions in the chamber and to be able to operate in high neutron flux is to choose the gas with a higher ionisation potential. With this respect, helium is the best candidate as its cross section for multiple ionisation is almost ten times smaller than argon or neon [4]. Nevertheless, He gas has an important diffuseness factor what enhances the probability of gas leakage during the operation of the chamber at high temperatures.

The combination of the results as above defines the main parameters of the SDFC to be operated in high intensity fluxes of some $10^{15}$ n.cm$^{-2}$.s$^{-1}$ with a relative error less than 0.25%: 4 mm external diameter, 0.5 mm gap, Ne or Ne+0.5%Ar at 1 bar and 20 $\mu$g $^{235}$U deposit.
3.3. Evolution in time

Because the chamber will be irradiated in a high thermal neutron flux for 50 days, the burn-up of $^{235}$U will be important. In addition $^{239}$Pu will be formed from 2.5% of $^{238}$U present in the initial composition of the deposit, and it will contribute to the saturation current. To determine this effect we have calculated the isotopic evolution of active deposit during the cycle of the ILL reactor using the evolution code CINDER [5]. Neutron flux in the experimental channel was calculated by the use of MCNP code. More details on these calculations can be found in Ref [6].

In Fig. 3, we can see that the current due to $^{239}$Pu is negligible (< 4 %) compared to the current due to $^{235}$U at the end of the cycle, leading to an error compatible with the accuracy of 6% required for the normal SDFC operation. This uncertainty can be easily removed by using an isotopically more pure $^{235}$U deposit.

![Figure 3: Evolution of the saturation current as a function of irradiation time in the HFR, with the following characteristics: 20 µg of $^{235}$U, the Ne gas at 1 bar and 0.5 mm for the gap.](image)

**Conclusions**

We show that the use of an innovative fission micro-chamber is possible for incineration experiments in high intensity thermal neutron fluxes. The operation in current mode and with specific filling gases will enable the development of double deposit fission chambers to measure on-line the incineration rate of minor actinides or specific actinide mixtures. This application in the frame of the Mini-Inca project opens a new field of application for these promising detectors. The Mini-Inca project and the neutron detection for the Mega-Pie project for an improved spallation target at the Paul-Scherrer Institute in Zurich are two examples.

**References**