Recent developments on micrometric fission chambers for high neutron fluxes

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Abstract— with the development of innovative nuclear systems and new generation neutron sources, the nuclear instrumentation should be adapted. Since several years, we developed microscopic fission chambers to study the transmutation of minor actinides in high thermal-neutron fluxes. The recent developments done to fulfill the drastic conditions of irradiations are described in this paper together with the feedback from the measurements. Two installations were used: the HFR of the ILL for its highest thermal neutron flux of the world and the MEGAPIE target which was the first 1 MW liquid Pb-Bi spallation target in the world.

Index Terms — fission chambers, transmutation, liquid spallation target, high flux reactor.

I. INTRODUCTION

Fission chambers are part of in-core or ex-core instrumentation in nuclear reactors. They are particularly well suited to monitor the reactor fuel burn-up and the start-up of the reactor due to their small size and a good signal-to-noise ratio. With the emergence of innovative nuclear systems for electricity production as proposed in the Generation IV initiative, or the development of intense neutron sources for various applications as material irradiations, nuclear waste incineration, etc, the question arises on how the nuclear instrumentation is adapted for these new devices. Indeed, these systems have in common a high level of neutron flux and for some of them high temperature. Moreover, when regarding neutron sources based on liquid spallation targets, magnetic perturbations have to be considered.

Within the framework of the Mini-Inca project [13] which is devoted to the study of minor actinide transmutation in high neutron fluxes and the MEGAPIE experiment [10] which was the first test of a 1 MW liquid metal spallation target, we started an instrumental program in collaboration with Photonis and CEA/DEN/DER/SPEX to develop and test micrometric fission chambers for neutron flux monitoring and transmutation studies in high fluxes. These detectors should sustain the drastic conditions of irradiation met in the two installations we used: the High Flux Reactor (HFR) of the Laue-Langevin Institut (Grenoble, France) and the MEGAPIE target at the Paul Scherrer Institut (Villigen, Switzerland). Among these conditions, one is the functioning in high burn-up conditions, where structural pieces are strongly activated, and the deposit is strongly damaged. Another constraint is the high temperature met in the MEGAPIE target and the fast component of the neutron flux which creates also damages in the materials.

Here is a brief history of the work done. Starting with the 4.7 mm fission chambers commercialized by Photonis (CFUF43), we saw that the main degradation when increasing the neutron flux from $10^{14}$ to $10^{15}$ n/cm$^2$/s was the disappearance of the so-called “plateau” in the measured current. As a consequence we suspected to lose also the proportionality to the generated primary electrons, i.e. to the neutron flux. Then we pursued our developments with the idea to recover this “plateau”. As we identified the accumulation of charges in the inter-electrode gap as being probably the main phenomenon responsible for this degradation we focused our developments on its reduction. All these developments were supported by strong modelisation efforts attempting to understand, at least qualitatively, the influence of the different physical processes involved in the current generation and responsible for its degradation. In parallel, we also developed a concept of compensated “back-to-back” fission chambers to follow on-line the incineration of Minor Actinides. This concept of detector is also well suited to monitor the thermal versus fast neutron component by using threshold fission reactions as $^{242}$Pu(n,f), for instance.

In the present paper we detail all these developments and for each modification we tested the feedback from experiences. It is divided into three parts. The first part describes the installations and their neutronic performances, the second and the third parts detail the specific developments for each installation.
II. DESCRIPTION OF THE EXPERIMENTS

A. The High Flux Reactor

The High Flux Reactor of the Laue Langevin Institut (Grenoble - France) is the highest neutron source in the world for thermal neutrons. It is composed of a single fuel element, containing 9 kg of highly enriched Uranium. It is cooled and moderated by a flow of heavy water. For our studies, we used the Vertical V4 channel which was rehabilitated in 2002 [12]. It is composed by two concentric tubes (internal diameter: 28 mm) introduced inside a tube inclined at 8°30’ with respect to the vertical. The channel is located inside the D\textsubscript{2}O reflector with its extremity distant of 360 mm from the core axis, in the median plan. Due to its close proximity from the core it offers a thermal neutron flux amounting to around $10^{15}$ n/cm\textsuperscript{2}/s. Moreover, due to its inclination, the neutron flux reduces when moving from the bottom (position 0 cm) to the top of the tube (see Fig. 1). The same goes for the epithermal component. The latter amount to about 15% of the total neutron flux at the lowest position and becomes negligible above 75 cm. For each irradiation, fission chambers were inserted in an irradiation cane and were maintained in a He gas environment.

The neutron fluxes were estimated from a precise modelling of the ILL reactor geometry with the MCNP2.5 code. The simulations were performed with a statistical accuracy better than 1% corresponding to high statistic events ($5\times10^6$ to $20\times10^6$ fission neutrons). The resulting neutron densities are shown on Fig. 2 for the 5 most widely used irradiation positions.

For all the measurements we used the 1 mm in diameter coaxial cables from Thermocoax containing alumina as electrical insulator. These cables have high resistivity and low sensitivity to radiations. The currents were measured by pico-ampmeters (Keithley 6487) and the readout ensures by a Labview program via GPIB connection.

B. The MEGAPIE target

The MEGAvatt Piplot Experiment (MEGAPIE) project was started in 2000 to design, build and operate a liquid metal spallation neutron target at the power level of 1 MW. The project is an important step in the roadmap towards the demonstration of Accelerator Driven System (ADS) concept and high power molten metal targets in general. With the 600 MeV cyclotron delivering a continuous beam with a current up to 1.4 mA, SINQ is the most powerful spallation neutron source in the world, approaching a proton beam power on target of 1 MW. In the MEGAPIE target a loop of about 82 liters lead-bismuth eutectic (LBE) circulated enclosed by a steel structure. The target was about 5 m long and the LBE was made circulating by means of a main electromagnetic pump, while a bypass pump was used for a second loop to cool the window [10].

One innovation introduced in the MEGAPIE experiment was the measurement and the monitoring of the neutron flux in the close proximity of the spallation zone inside the target. We built an instrumented cane containing 8 micrometric

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Fig. 2: Neutron flux energy spectra as simulated with MCNP2.5 code at different irradiation positions in the V4 channel. Also are indicated the flux densities in the core and in the H9 channel. The integrated flux intensity varies from $6\times10^{13}$ n/cm\textsuperscript{2}/s at 100 cm to $1\times10^{15}$ n/cm\textsuperscript{2}/s at 0 cm.

Fig. 1: Schematic view of the bottom part of the instrumented cane. Not to scale.
fission-chambers distributed by pair and 9 neutron flux monitors (see Fig. 1). The lowest stage was shielded with metallic Gd foils, making the detector at this position only sensitive to epithermal neutrons (see Fig. 5). The cane was 4.7 m long, 12 mm in diameter in its lower part and 22 mm in the upper part. The cane was inserted in the central rod. A more detailed description of the experiment can be found in [2].

The neutron fluxes were estimated from a precise MCNPX2.5.0 simulation taking into account the detailed geometry of the MEGAPIE target and the SINQ facility it is inserted, including the D$_2$O moderator. On Fig. 5 are shown the neutron flux density as “seen” by the fission chambers.

III. DEVELOPMENT OF FISSION CHAMBERS FOR HFR

A. Functioning of fission chambers in high neutron fluxes

Fission-chambers are ionisation detectors containing fissile elements. In our case, the geometry is cylindrical (see Fig. 4) and the fissile element is electrodeposited onto the outer surface of the anode. The inter-electrode gap is filled with pressurized gas, in our case Argon at about 1 bar. When fission occurs, a signal is generated by one fission product which ionises the internal gas. Two modes of functioning are usually used: the pulse mode and the current mode. In the pulse mode, each pulse generated by the collection of electrons and ions is recorded.

But when the fission rate increases, pulses pile up. It is then possible to continue with this mode by using a statistical treatment of the signal in the so-called Campbell mode or fluctuation mode. If we only consider the first moment of this mode, we are in the current mode. In this mode any statistical information that can be used to reject for example noise is lost. Then the background current has to be measured separately with a fission chamber without fissile deposit. The advantage of such measurement is to include the parasitic currents generated by electromagnetic radiations, in particular the $\gamma$-rays emitted by the installations, and the activation of pieces. In the following we only worked in the current mode, accompanying each fission chamber with a chamber without any deposit for background subtraction.

We based our developments on the 4.7 mm in diameter fission chambers (CFUF43) commercialized by Photonis. These small gap detectors are well adapted for high neutron fluxes. On Fig. 3 are shown the measured currents as a function of the applied voltage for the different irradiation positions in the V4 channel. In the highest position where the neutron flux is below $10^{14}$ n/cm$^2$/s we clearly recognise the three domain description of a fission chamber: a recombination domain below 120 V, a “plateau” between 120 V and 200 V, and the “avalanche” domain at higher voltage. In the recombination domain, the primary electrons generated by the ionisation of the Ar gas by fission products are not fully collected by the anode. They are absorbed by positive ions drifting inside the inter-electrode volume. In the “avalanche” region, primary electrons have sufficient energy to produce secondary electrons close to the anode, enhancing the collected current. Between these two domains, in the “ionization chamber” mode, the applied electric field is high enough to prevent recombination, and not high enough to induce secondary electrons. In this domain, we estimate that all primary electrons and ions are integrally collected, thus the measured current is directly proportional to the fission rate and when corrected from the effective fission cross section, to the neutron flux.

When increasing the neutron flux we see that the “plateau” vanishes and tends to an inflexion point. We could estimate
this transition to be around $2 \times 10^{14}$ n/cm²/s. In [3] and latter on [4] we have studied the origin of this effect. In the following we only highlight on the main conclusions. The physics of a fission chamber can easily be described by the basic charge transport equations to describe the evolution of electron ($\rho_e$) and ion ($\rho_i$) densities:

$$\frac{\partial \rho_e}{\partial t} + \text{div}(\rho_e v_e) = T_{\text{SOURCE}}^e + T_{\text{LOSS}}^e,$$

$$\frac{\partial \rho_i}{\partial t} + \text{div}(\rho_i v_i) = T_{\text{SOURCE}}^i + T_{\text{LOSS}}^i,$$

where the $T_{\text{SOURCE}}$ and $T_{\text{LOSS}}$ are the creation and disappearance terms, and from the Poisson equation to describe the electrical field ($E$) generated by a difference of electrical potential ($V$):

$$\text{div}(E) + \frac{\rho_e - \rho_i}{\varepsilon_0} = 0,$$
$$\int E \, dl = \Delta V.$$

In current mode (1) are stationary state equations so that the time derivative terms disappear. The creation term $T_{\text{SOURCE}}$ is mainly the sum of two contributions. First, a term $T_{\text{SOURCE}}^1$ due to the ionization of the gas by the fission products. It only depends on the fission rate, the effective ionisation potential of the gas and the geometry of the chamber. Secondly, a term $T_{\text{SOURCE}}^2$ resulting from secondary ionizations due to electrons which have acquired in the electric field enough kinetic energies. It is usually written

$$T_{\text{SOURCE}}^2 = \alpha n_v v_e,$$

where $\alpha$ is the Townsend first ionization coefficient and $v_e$ the drift velocity of electrons.

The disappearance term $T_{\text{LOSS}}$ is mainly due to the recombination of electrons with ions. These processes of capture proceed in three steps, classified by size scale:

- Initial: the ejected electron recombines with the ion from which it comes from. This mode of recombination plays only at high pressures (10 to 100 atmospheres).
- Columnar: the pairs can then recombine inside the ionized tracks left by the fission products in gas. These tracks have indeed high densities of charges.
- Voluminal: the most general process. The electrons migrate in gas under the effect of the electric field and can recombine with any ion they meet.

For pressures close to the atmosphere, the dominating process is the voluminal recombination. $T_{\text{LOSS}}$ can thus be written as follows

$$T_{\text{LOSS}} = kn_v n_e,$$

where $k$ is called the “recombination coefficient” and depends on the nature of the gas.

By solving (1) and (2) with some simplifications, it was shown in [3] that when increasing the fission rate, the saturation “plateau” gradually disappears and the transition between the recombination and the avalanche domains is less and less pronounced. This major perturbation is primarily due to the reinforcement of the recombination term $kn_v n_e$, the charge density increasing with the fission rate. In addition, it was shown that the measured current at the inflexion point does not vary linearly with the fission rate above about $10^9$ fissions/s.

![Fig. 6: Calculated distortion of the electrical field induced by space charges as a function of the radial distance r for different applied voltages from [3].](image)

Indeed, due to the limited drift velocity of electrons and particularly of ions in the gas, charges can accumulate in the inter-electrode volume if their production rate is larger than the time needed to collect them. As consequences, the density of ionized atoms increases in the volume, increasing by the way the recombination with primary drifted electrons and modifying the electrical field between the two electrodes (see Fig. 6). The space charges induce a diminution of the electric field at the anode and an increase at the cathode. This field distortion is reinforced when the voltage decreases, increasing the collection time. Thus the recombination domain extends to higher voltages and the avalanche domain start at lower voltage, explaining the distortion in the current response of the detector when increasing the neutron flux.

**B. Developments for high neutron fluxes**

Then following the theoretical work of [3] and later on [4-7] we have focused our developments on the comprehension of the functioning of fission chambers in high fluxes and on the way to reduce the accumulation of charges in the inter-electrode volume. We tried to play on several parameters: the mass of the fissile material, the geometry of the detector and the pressure and the nature of the gas.
1) Mass of the deposit
A parameter on which it is easy to play is the reduction of the initial quantity of fissile deposit. We gain a factor 10 on the $^{235}$U mass, reducing by the way the fission rate by a factor 10 and the charge effects. Unfortunately, the electrolyze method we used does not allow us to go down below few micrograms, principally due to the question of homogeneity of the solution. The reduction of fissile atoms is probably a promising way to explore but also limited by the ratio of the signal over background. To give an idea, with 4 $\mu$g of $^{235}$U at the beginning of irradiation, the signal over background is about 2-3 (see Fig. 7). By the end of irradiation (after 50 days) the ratio becomes below 1.

2) Geometry of the detector
From [3] it was shown that by reducing the inter-electrode gap we reduce the charge effects. The basic ideas are first to reduce the creation of electron-ion pairs and then to reduce the time of charge collection due to the proximity of the electrodes. In [8] this option was already explored by reducing the cathode diameter to 1.5 mm.

We choose the solution to keep the same cathode diameter and to increase the anode radius reducing the gap from 500 $\mu$m to about 250 $\mu$m (CFUT-C6). The main technological challenge was to maintain the anode axis as close as possible from the cathode axis to avoid electrical perturbations. We solve this problem by fixing the two extremity of the anode. In this configuration it was shown in [3] that the electrical distortion should be reduced by at least 1 order of magnitude due to a reduction of the charge space by a factor 13. On Fig. 8 are shown the resulting (I/V) curves for CFUT-C6 irradiated at 25cm. We observe that the measured current shows a “plateau” at the beginning of irradiation even in a neutron flux of $8 \times 10^{14} \text{n/cm}^2/\text{s}$. The recombination domain is still important and as expected reduces when the neutron flux reduces whereas the avalanche domain seems to be independent of the flux. Unfortunately, we see also a degradation of the fission signal with the time elapsing: the avalanche domain increases whereas the recombination domain decreases. The decrease in the recombination domain is due to the burn-up of the fissile element as it seen in position 75 cm where we clearly see that the fission chamber is still well functioning. The increase of the avalanche domain is much more difficult to explain but it is probably due to the presence of impurities formed during the irradiation. The effect is much more pronounced in high fluxes than in lower fluxes.

The presence of impurities is effectively one of the main problems for the degradation of the signal in high fluxes. It could be reduced but not avoided. Indeed even if we prevent from the degassing of structural pieces under irradiation due to the $\gamma$-heating and neutron induced-damages, there are still volatile fission products which are generated and could be responsible for a pollution of the mono-atomic gas. Another source of pollutant could be the sputtering of the metallic cathode when submitted to an intense bombardment by the fission products. This phenomenon depends on the metal choose for the cathode and the quality of its surface. Finally the deposit itself can generate impurities, in particular oxygen when it is an oxide form. The possibility to use deposit under the metallic form should be explored.

All these contaminants have lower ionisation potential than noble gases and consequently reduce the effective ionisation potential. One of the well known processes responsible for such reduction is the Penning ionisation: the energy contains in an excited (metastable) state of the noble gas is sufficient to
ionize the impurity when interacting.

We improve the fabrication process by systematically degassing all pieces, including the deposit, at least one night at a temperature of 400°C before to assemble them. This has clearly improved the quality of the signal, especially during the first hours of irradiation. In [8] it is proposed to use heavier noble gas to reduce the impact of impurities. This solution has to be tested.

3) The gas parameter

In [1] it is shown that the use of He or Ne gases is better than Ar to go to higher neutron flux. This conclusion was mainly based on the ionisation potential which is much higher for He and Ne gases than Ar. In consequences, the number of electron-ion pairs should be reduced when using these gases. When we tested a fission chamber filled with Ne at 1 bar, the saturation plateau was invisible whereas it was present in the response of the fission chamber filled with Ar at 1 bar. One possible explanation was later on given in [3] by the so-called Jesse effect: in presence of impurities the number of pairs is enhanced in gases having high ionisation potential (He or Ne).

We also tested the pressure of the gas (Fig. 7). The risk when reducing the pressure is to create sparking in the inter-electrode volume degrading by the way the deposit. For this reason, we never tried to go down below 0.8 bars. We saw that more or less the signal follows the Ar pressure: higher the pressure is, higher is the signal. We never went above 1.2 bars.

We also removed the external insulation. In [12] each chamber was surrounding with alumina to insulate electrically each others. Unfortunately the electrical independence of the two chambers was not perfect resulting in a cross talk between the two signals. One of the explanations was that the ionized gas could communicate via the 1 mm gap in the cap. In the following generations of chambers (CFUT-C7) we solved this problem by reducing the gap in the cap. We also used the external insulation.

The first concept, consisting of two fission chambers connected altogether by the cap in which the gas could circulate freely in a gap of about 1 mm, was tested in 2002 [12]. Each chamber was surrounding with alumina to insulate electrically each others. Unfortunately the electrical independence of the two chambers was not perfect resulting in a cross talk between the two signals. One of the explanations was that the ionized gas could communicate via the 1 mm gap in the cap. In the following generations of chambers (CFUT-C7) we solved this problem by reducing the gap in the cap. We also removed the external insulation.

The third chamber was added to measure the current which is not induced by fission. It is essentially generated by the interaction of γ-radiation on materials generating electrons in the gas and current in the cables. A small part is also due to electrons generated by the β-decay of activated pieces. In the lowest position of V4, where the γ-flux is more or less the same than the neutron flux, it amounts to about 30 μA whereas at 75 cm it only amounts to 4 μA (see Fig. 9).

The fission current is then deduced from the measured current by subtracting this measured background current and can be expressed as:

\[ I_{\text{fission}}(t) = I_{\text{measured}}(t) - I_{\text{background}}(t) = \Gamma N_0 \hat{\sigma}_{\text{fission}} \phi(t) f(t), \]

where \( \Gamma \) is the sensitivity factor of the detector which depends only on the geometry and the gas properties (identical for all chambers), \( N_0 \) the initial atom quantity (control at the level of 1% by mass spectrometry), \( \hat{\sigma}_{\text{fission}} \) the effective fission cross section, \( \phi(t) \) the neutron flux and \( f(t) \) contains the evolution information of the actinide. At the beginning of irradiation \( f(0)=1 \) and the fission current should be linear with the reactor power. Fig. 11 shows this linearity during the start-up phase of the reactor for a CFUT-C7 irradiated at 25 cm and showing the good response of the detectors.

C. Fission chambers for transmutation studies

For transmutation studies we needed detectors able to function at high burn-up and allowing the measurement of fission rates in a close place. We developed a concept of compensated fission chambers (CFUT-C7), based on a modified concept of the “back-to-back” fission chambers, to measure online the evolution of the fission rate of a fertile isotope in reference to the fission rate of 235U. By this way, fission cross sections but also capture cross sections could be extracted from the evolution of the measured fission current (Fig. 9 shows the latest measurement to extract the fission and capture cross sections of 238Pu). It consists in three fission chambers electrically independent but sharing the same Ar gas via the cap (see Fig. 10).

![Fig. 10: Photography of a triple deposit fission chamber (CFUT-C7).](image)

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On Fig. 12 is shown the \((I/V)\) curve for the \(^{238}\text{Pu}\) chamber associated to Fig. 9. An interesting point is that even at \(3 \times 10^9\) fissions/s, the plateau still exists and the curve does not degrade too much with time whereas we have seen on Fig. 8 a strong deformation for the same fission rate. It confirms the hypothesis that the observed degradations for CFUT-C6 are probably due to the formation or the presence at the beginning of impurities. Between CFUT-C6 and CFUT-C7 we also improved the filling procedure taking care to remove all impurities before filling with Ar.

IV. FISSION CHAMBERS FOR MEGAPIE

A. Specific developments

One of the challenges for the MEGAPIE experiment was to develop detectors for high temperature. During the functioning the Pb-Bi liquid was maintained at a temperature of 250°C and the temperature was increasing to 400°C with the beam on. Moreover, during the filling phase of the target with Pb-Bi, the entire target was heated to 500°C. Thus the specifications for the global detector were:

- to stand temperature up to 600 °C;
- to stand to a water pressure of 1.6 MPa;
- to be insensitive to electromagnetic fields;
- to measure thermal and fast neutrons.

The main difficulties when developing a detector for high temperature is to guarantee the mechanical integrity of the detector, the electrical contact and to minimise the degradation of the resistivity. The choice of the materials, the conception and the welding were very important to insure the mechanical integrity and in particular to avoid differential dilatation between pieces in contact. By choosing the stainless steel (304L) and titanium (TA6V) for structural pieces and pure alumina as insulating material we guaranteed the mechanical integrity and insulation of the fission chambers up to 600°C. The electrical contacts were reinforced when necessary with Inconel springs tested up to 600°C for their elasticity. Different cables from Thermocoax were also tested and improved to fulfil the requirements [2].

Another point was concerning the behaviour of the deposit under temperature cycling. This question was investigated by means of Scanning Electron Microscopy analysis after having cycling in temperature up to 600°C the uranium deposit. As shown on Fig. 13 the deposit is still stick on the anode and the surface is even cleaner.
- micrometric fission chambers are well suited for high neutron fluxes. We tested commercialized fission chambers to work perfectly up to $2-3 \times 10^{14}$ n/cm²/s in current mode and the modify ones up to 400°C.

- the prominent restricting parameter is the accumulation of charges within the inter-electrode gap. We tried to limit their accumulation by reducing the inter-electrode gap and the mass of the deposit which seems to be a promising way.

- another restricting parameter is the purity and the quality of the mono-atomic gas during the fabrication and the irradiation that have to be preserved to ensure the functioning of the detector. Solutions have to be found and strong efforts have to be supplied to the fabrication process in order to keep as clean as possible all pieces and to remove all the degassing processes.

Finally, we developed a concept of triple deposit fission chambers for transmutation studies that can be used also to get the thermal over fast component ratio of the neutron flux.

**ACKNOWLEDGEMENT**

We would like to thanks the technical staff of the ILL reactor division for their help and support.

**REFERENCES**


