A compact fission detector for fission-tagging neutron capture experiments with radioactive fissile isotopes


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was designed aiming at reducing the background from the detector and the goal of measuring the background due to its large scattering and capture cross-sections. With such a detector is made out of copper, which is a significant source of micro-mesh. In the recent measurements performed at n_TOF a Micromegas based detector was used as a fission detector [2,4]. The so-called γ-veto technique. In addition to the background has been developed, the so-called fission-tagging or fission-veto technique. In the past [1] a method of efficiently dealing with this source of background, which has to be taken care of in the analysis. However, the knowledge of those cross-sections is limited due to difficulties associated to the background from the fission reaction channel. For the fissile isotopes $^{233}$U, $^{235}$U and $^{239}$Pu the fission cross-section is on average a factor 2 to 10 larger than their respective capture cross-section, depending on the isotope and energy range, which is shown in Fig. 1. This implies that in a measurement of the capture cross-section the γ-rays coming from the fission channel are a major source of background, which has to be taken carefully of in the analysis. In the past [1] a method of efficiently dealing with this source of background has been developed, the so-called fission-tagging or fission-veto technique. In addition to the γ-detector this technique employs a fission detector to measure the fission fragments. The γ-rays from the fission reaction can then be identified or tagged by operating the two detectors in coincidence. In recent years new efforts have been made to measure the capture cross-sections of the fissile isotopes using the fission-tagging technique at different facilities [2–5]. Despite fission-tagging technique’s effectiveness in dealing with the fission background, it has the drawback of introducing another component to the background, namely the sample substrates and the detector itself. In the recent measurements performed at n_TOF a Micromegas based detector was used as a fission detector [2,4]. The so-called micro-mesh of such a detector is made out of copper, which is a significant source of background due to its large scattering and capture cross-sections. With the goal of measuring the $^{233}$U(n, γ) cross-section a new fission detector was designed aiming at reducing the background from the detector and providing the necessary performance to reliably identify the fission γ-rays. The design of the described fission chamber (FICH) is adapted to the measurement of the $^{233}$U(n, γ) utilizing the fission chamber coupled to the n_TOF Total Absorption Calorimeter (TAC) [6] in experimental area 1 (EAR1) of the n_TOF facility [7].

2. The multi-plate fission chamber

Due to the experience obtained from the measurement with the fission-tagging micromegas detectors [2,4], a simple ionization cell geometry is chosen as the basic detector design to minimize the material in beam. The development of the fission chamber is focused on different, partially contradictory, criteria: excellent time resolution for the coincidence and time-of-flight measurement; low quantities of structural material to avoid additional background for the capture measurement; reasonable amount of $^{233}$U to obtain a sufficient count rate for high statistics measurements; compact design as the fission chamber (FICH) must fit inside the TAC of n_TOF. All these requirements are detailed in the following sections.

2.1. Technical description

As a result the fission chamber (FICH) is designed as a multi-plate ionization chamber containing two stacks of axial ionization cells. Figs. 2 and 3 show CAD drawings and pictures of the chamber. The housing is made of a 1.5 mm thick aluminium tube with an outer diameter of 66 mm and a length of 78 mm. With a maximum outer diameter of 90 mm and a total length of 120 mm including the flanges with the gas connections and windows, it fits nicely in the γ-calorimeter leaving sufficient space for its absorber (explained in Section 3.1.2) and the connecting beam pipes. Two stacks of seven ionization cells
each are mounted directly on their respective motherboards and are inserted from each end of the chamber. The stacks have a minimum inner diameter of 50 mm leaving enough space for the n_TOF neutron beam with a FWHM of roughly 16 mm and a total width of less than 40 mm. In total 8 anodes are collecting signals from 14 $^{233}$U targets deposited on the cathodes. The arrangement of the cathodes, anodes and deposits is illustrated in Fig. 4. To avoid cross-talk from alpha-particles, the ionization cells are separated by 20 μm aluminium, either one 20 μm anode foil or two 10 μm cathode foils, resulting in a total of 300 μm aluminium in the neutron beam which is a negligible neutron beam perturbation. The chamber is closed with aluminized 25 μ thick Kapton windows to provide a Faraday cage.

2.2. Choice of gas and gas system

The gas is of high importance and has to exhibit a high drift velocity and provide the best possible alpha-fission separation. High purity tetrafluoromethane $\text{CF}_4$ is a fast gas and is often used where high count rates are expected [8] but has the drawback of being electronegative, worsening the energy resolution and hence the alpha-fission discrimination. Nevertheless, the advantage it offers due to its higher drift velocity compared to other gases outweighs the disadvantages. Fission fragments (FF) and α-particles deposit their energy in the gap between the electrodes filled with the gas. Simulations [8] have shown that a gap distance of about 1.5 mm is sufficient to achieve a reasonable alpha-fission separation for $^{252}$Cf. Due to mechanical considerations the gap is chosen to be 3 mm. To achieve a drift velocity of about 11 cm/μs an electric field of 1400 V/cm is applied at atmospheric pressure. This drift velocity corresponds to a total electron drift time of 27 ns in the 3 mm gap, leading to a suitable intrinsic time resolution. In order to guarantee stable conditions throughout the measurement period of four weeks a gas pressure and flow regulation system was employed and is schematically shown in Fig. 5. The fission chamber was operated with a constant gas flow of 0.1 l/min and at an absolute pressure of 1100 mbar to allow for the use of thin windows of the fission chamber, hence to reduce the background in the $\gamma$-calorimeter.
2.3. Dedicated electronics

To ensure a good time resolution and reduce potential $\alpha$-particle pile-up, fast electronics adapted to the geometry of the ionization cells and the electron drift velocity have been developed. Charge preamplifiers with a short RC decay time constant have been developed to ensure good amplitude discrimination, avoid saturation due to very high alpha activity and to preserve the good timing response of the chamber. A dedicated card combining the preamplifier and a fast timing filter amplifier was directly mounted on the fission chamber. This reduces the input capacitance and improves the signal-to-noise ratio. A picture of those cards can be seen in the lower left part of Fig. 5. The signals recorded by the data acquisition system were digitized, stored and processed offline using the pulse shape analysis routine developed by the n_TOF collaboration [9]. An example of a typical signal of a fission fragment is shown in Fig. 6 with a full width at half maximum (FWHM) of 34 ns and a rise time (10-90%) of 16 ns.

2.4. Fissile deposits

Thin uranium oxide layers, with 99.9361% enrichment in $^{233}$U, had a diameter of 40.00 ± 0.02 mm and were molecular plated on 10 $\mu$ thick aluminium foils at JRC-Geel. The impurities in the sample have a negligible effect on fission. Nevertheless, a small effect on the $^{235}$U $\alpha$-ratio is expected due to the first capture resonance of $^{234}$U at 5.15 eV. This contribution can be taken into account during the resonance analysis. The activity of each of the 14 samples hosted in the chamber has been determined by well-defined solid angle $\alpha$-particle counting and amounts to an average $\alpha$-activity of about 1.16 MBq or an average...
areal density of 264.5 μg/cm² per sample (with a standard deviation of 30.9 μg/cm² among the 14 samples), which permits fission fragments to escape the deposits, resulting in a total mass of 46.5(3) mg of 233U.

3. Fission-tagging experiment at n_TOF

233U is a prime example for the application of fission tagging as it exhibits a fission cross-section which is on average a factor 10 larger than the corresponding capture cross-section. Thus, the fission reaction will introduce a background into the measurement that comprises of two components: the prompt component caused by the de-excitation of the highly excited fission products and the delayed component caused by either fission neutrons being captured in the experimental set-up or decays of unstable fission fragments with half-lives larger than a few nanoseconds up to microseconds. The prompt component causes a much larger background and appears quasi-instantaneous with the fission reaction and can be easily quantified and removed using fission-tagging. The delayed component can also be studied with fission-tagging but depends on the experimental set-up's sensitivity to neutrons and shall not be the focus of this work.

3.1. Experimental set-up

3.1.1. The n_TOF facility at CERN

The n_TOF experimental area 1 (EAR1) facility [7] is devoted for the measurement of energy dependent neutron cross-sections in an energy range from thermal up to GeV. Neutrons are produced by a high-intensity 20 GeV/c proton beam impinging on a lead target and moderated in a borated water-layer down to thermal energies. The proton beam is delivered by CERN's Proton Synchrotron with an average proton beam intensity of 7⋅10^12 or 4⋅10^12 protons per bunch for dedicated or parasitic bunches respectively. The neutron fluence as a function of the arrival time in EAR1 located approximately 185 m from the lead target is shown in Fig. 7.

The n_TOF facility provides a fully digital Data Acquisition system (DAQ) [10] and a large storage space, namely the CERN Advanced STORage manager (CASTOR) [11]. The waveforms of all signals are digitized with high performance digitizers, ADQ412 or ADQ414 [12], with 12 or 14 bit resolution respectively which are operated at 500 MSamples/s. This allows an offline analysis to be performed with dedicated pulse shape analysis routines [9]. The digitizers are triggered with a common external clock to avoid time drifts between the different channels.

3.1.2. The n_TOF Total Absorption Calorimeter

The n_TOF Total Absorption Calorimeter TAC [6] is designed to detect in coincidence the γ-rays of the electro-magnetic cascade following a neutron capture event. The TAC is a segmented 4π scintillator array consisting of 40 BaF₂ crystals mounted in a honeycomb structure which holds the full spherical detector shell as shown in Fig. 8. The spherical BaF₂ shell has a 20 cm and 50 cm inner and outer diameter respectively, covering 95% solid angle resulting in an efficiency of detecting at least one γ-ray from a cascade close to 100%. To reduce the neutron sensitivity, namely the probability of detecting neutrons of the beam scattered from the in-beam materials, a so-called absorber is placed between the crystals and the sample to be measured. The absorber is made out of polyethylene loaded with 7.56 wt% natural lithium to absorb scattered neutrons and consists of two spherical shell halves in which the fission chamber was embedded as shown in the right panel of Fig. 3. TAC events are characterized by three parameters: the time-of-flight, the number of hit crystals referred to as crystal multiplicity $m_{cr}$ and the sum of the deposited energy in all 40 crystals $E_{Sum}$ within a time coincidence window of $T_{TAC coincidence} = 12$ ns.

3.2. FICH Performance

3.2.1. Pulse height spectrum and alpha-fission discrimination

Fig. 9 shows the pulse height spectrum of the fission chamber for neutrons of less than 10 keV energy and without neutron beam (beam off). Small pulse heights are dominated by the α-particle background and are several orders of magnitude larger than the contribution of the

3.2.2. Neutron fluence at n_TOF EAR1 185m from the source.
fission events. The blue line in Fig. 9 is a scaled version (for visualization purposes) of the pulse height spectrum of the fission chamber without neutron beam and shows the $\alpha$-peak which corresponds to $\alpha$-particles that deposit their full energy in the gas.

The relatively poor separation of fission fragments and $\alpha$-particle background at around 0.09 V is not surprising considering the high $\alpha$-particle count rate. Choosing appropriate conditions allows to study the response of the FICH to fission fragments with a much better separation as shown in Fig. 10 where the pulse-height spectra for different $E_\alpha$ are displayed. For example, gating on the first and largest resonance at $1.6 \text{ eV} < E_\alpha < 1.9 \text{ eV}$, corresponding to the TOF region of 10.6-9.7 ms, improves the separation of $\alpha$-particles and fission fragments. Furthermore, the characteristics of the neutron fluence at n_TOF EAR1 can also be exploited by choosing the neutron energy region around of corresponding to a TOF range of 5-15 $\mu$s (0.8-7 MeV), see Fig. 7. Due to the higher fluence in this TOF region the alpha-fission separation is further improved.

### 3.2.2. Gain monitoring

The gain of the FICH has been monitored throughout the measurement by counting the number of fission fragment (FF) events (> 0.1 V) per nominal ($7 \cdot 10^{12}$ protons) pulse. Fig. 11 shows the gain fluctuation of one of the ionization cells over time, indicated in RunNumber. No drift of the gain can be observed, proving a good detector stability throughout the whole measurement time of about four weeks.

### 3.2.3. Dead time and validation

With high count rates dead time and pile-up can become severe. Due to its design as a fast fission chamber, count rates of several MBq should be sustainable without the need to correct for pile-up effects in fission fragment detection. Fig. 12 shows the ratio of the count rates for dedicated (D) and parasitic (P) beam pulse types of fission events ($\text{amp} > 0.1 \text{ V}$). A good agreement with 1% is reached up to 1 MeV, indicating that there are no pile-up or dead time issues. The outlier around 55 keV most likely corresponds to dips in the neutron flux due to aluminium resonances, hence very low statistics.

To verify the satisfactory behaviour of the fission detector the shape of the $^{233}\text{U}(n,\alpha)$ cross-section has been calculated from the FICH events and the shape of the neutron flux. The resulting shape of the $^{233}\text{U}(n,\alpha)$ cross-section has then been normalized to evaluated libraries in the neutron energy range from 8.1 eV to 17.6 eV because this region is well separated avoiding interference from neighbouring resonances, as has been suggested in [13]. Fig. 13 shows the ratio of the scaled $^{233}\text{U}(n,\alpha)$
cross-section obtained from this work and the evaluated libraries, ENDF/B-VII.1 [14], ENDF/B-VIII.0 [15], JEFF-3.3 [16] and JENDL-4.0u2 [17], from 0.1 eV up to 10 keV. The deviations are within the evaluations’ uncertainties in the resolved resonance region (<600 eV) while the evaluations are discrepant in the unresolved region (>600 eV). Thus, taking only the resolved resonance region into account it can be concluded that the fission chamber is working satisfactorily in the neutron energy range of this measurement (<10 keV). An accurate prompt fission background subtraction for the measurement of the 235U(n, γ) cross-section can thus be assured.

3.3. Fission tagging

Events that produce signals in both detectors (FICH & TAC) in coincidence are related to fission events. The time correlation is given by the time difference between the detection of the event in the two detectors. Prompt fission events (small time difference) are characterized by high γ-multiplicity [18], as was observed and suggested in previous works [2,3].

3.3.1. Event reconstruction

The coincidence algorithm is based on the use of a coincidence window $T_{TOF_{TAC} - TOF_{FICH}}$ between TAC and FICH and allows positive and negative time differences. Fig. 14 shows the distribution of time differences $δT = TOF_{TAC} - TOF_{FICH}$ for all found coincidences and can be explained as follows:

- Events with $δT < -200$ ns show a flat distribution and correspond to random coincidences.
- The shape for $-200$ ns < $δT$ < 20 ns can be described by an exponential sitting on top of the constant background. The exponential increase corresponds to events where a γ-ray is emitted before the nucleus fissions. These events can be explained by the existence of the (n, γ) process (fission isomers) [19–22].
- A main peak for $10$ ns < $δT$ < 10 ns corresponding to the prompt fission events as suggested by the characteristics of those events with high $E_{sum}$ and $m_c$, indicated by the blue line.
- Another sharp structure or side peak for $10$ ns < $δT$ < 20 ns is an artefact of the event reconstruction process. The time difference between the main peak and this side peak corresponds exactly to the TAC coincidence window of 12 ns which is the minimum time difference between two TAC events due to how the TAC coincidence reconstruction algorithm works. The position of the side peak will shift with the TAC coincidence window.

Events with $δT > 20$ ns form an exponential tail and correspond to delayed events. Such events can be induced by fission neutrons which are subsequently captured in the experimental set-up thus emitting γ-ray cascades or isomeric states of the fission products that de-excite via γ-ray cascades with a delay corresponding to the half-life of the isomeric state. These events are related to fission but are not prompt fission γ-rays.

For reasons of causality the TAC-FICH coincidence window may not be smaller than the TAC coincidence window, otherwise there is the possibility of losing coincidences artificially. The optimal time window is a compromise between pile-up and efficient tagging. $T_{TOF_{TAC} - TOF_{FICH}} > T_{TOF_{TAC} - TOF_{FICH}}$ can lead to multiple coincidences found for a single FICH event. The different coincidences will be characterized by different TAC events, hence different $E_{sum}$, $m_c$ and $δT$. If two or more TAC events are assigned to the same FICH event, the TAC event with the highest crystal multiplicity $m_c$ is selected as the corresponding prompt fission event. If the TAC events happen to have the same crystal multiplicity then the event with higher $E_{sum}$ is selected as the corresponding prompt fission event. In principle these criteria are arbitrary and the performance of the different event selection algorithms is illustrated in Fig. 15. It shows...
the $E_{\text{sum}}$ spectra of the corresponding prompt fission events selected with different algorithms. It is evident that no matter which algorithm is chosen the difference is negligible.

Fig. 16 shows the effect of different coincidence windows $T_{\text{TAC-FICH}}$ on the total number of found coincidences normalized to the total number of fission events detected by the FICH (black dots; left axis). A steady increase can be seen with increasing $T_{\text{TAC-FICH}}$ which is understandable, although those additionally found TAC events in coincidence are not necessarily related to the prompt fission event but might correspond to random or delayed events. On the other hand Fig. 16 shows the number of coincidences where exactly one TAC event is found for one FICH event (red squares; right axis). With increasing coincidence window $T_{\text{TAC-FICH}}$ the number of one to coincidences drastically decreases, as the probability of multiple tagging starts to increase. A coincidence window $T_{\text{TAC-FICH}}$ slightly larger than the $T_{\text{TAC}}$ is already sufficient to tag close to 99% of the FICH events while a window too large might result in an uncertain assignment of multiple TAC events to a FICH event. To reduce this uncertainty the $T_{\text{TAC-FICH}}$ coincidence window is set to 14 ns.

FICH-TAC coincidence tagging also allows for a better alpha-fission separation as the probability of tagging an $\alpha$-particle is negligible compared to a fission fragment. In Fig. 17 the tagged fission amplitude spectra for different $E_n$ regions are compared to the amplitude spectra of the best achievable separation solely using the FICH. The improvement is obvious and allows the investigation of the shape of the fission fragment energy deposition in the fission chamber below what was possible with the FICH alone. It shall be noted that the TAC data for 0.8 MeV < $E_n$ < 7 MeV is usually not used in the analysis of cross-sections due to the so-called $\gamma$-flashes effect [6,7,23] which blinds the detector.

### 3.3.2. Tagging and FICH efficiency

In analogy to previous works [2,4], the tagging efficiency $\varepsilon_{\text{TAG}}(A_\text{th}; E_{\text{sum}}, m_{\nu})$ describes the probability of detecting a fission event identified as such by the FICH in the TAC and depends on the applied amplitude threshold $A_\text{th}$. It is defined as the ratio between the tagged fission events $c_{\text{TAG}}(A_\text{th}; E_{\text{sum}}, m_{\nu})$ and the total fission events detected by the TAC $c_{\text{TAC}}(E_{\text{sum}}, m_{\nu})$ (dependencies on $E_{\text{sum}}$ and $m_{\nu}$ are implicit for readability):

$$\varepsilon_{\text{TAG}}(A_\text{th}; E_{\text{sum}}, m_{\nu}) = \frac{c_{\text{TAG}}(A_\text{th}; E_{\text{sum}}, m_{\nu})}{c_{\text{TAC}}(E_{\text{sum}}, m_{\nu})}$$  \hspace{1cm} (1)
Fig. 18. Fission tagging efficiency $\epsilon_{\text{Tagg}}$ as a function of the crystal multiplicity and sum energy for two different amplitude thresholds $A_h$ in the neutron energy interval from 1.6 eV to 1.9 eV.

Fig. 19. Fission tagging efficiency calculated in several neutron resonances for $A_h = 0.076$ V, $m_c > 6$ and $E_{\text{Sum}} > 10$ MeV, their weighted average (red line) and the standard deviation of the data points (blue dashed lines). The uncertainties are calculated from the two highly correlated quantities in Eq. (1).

Fig. 20. Tagging efficiency $\epsilon_{\text{Tagg}}$ as a function of the FICH amplitude threshold (black circles). A scaled FICH amplitude spectrum in coincidence with the TAC for events with $m_c > 6$ and $E_{\text{Sum}} > 10$ MeV is shown too (grey line) as it is directly related to the efficiency.

4. Conclusions

A new compact fission chamber was developed and optimized for the use in fission tagging experiments to measure capture cross-sections of fissile isotopes. The development aimed at the use of the detection system at the n_TOF facility (CERN), coupled to the Total Absorption Calorimeter of EAR1, but can be generalized to other set-ups. The fission chamber was optimized for timing performance with an average signal rise time of about 16 ns and a FWHM of 34 ns which is optimal for the high specific $\alpha$-particle count rates from $^{233}$U as well as the alpha-fission discrimination and allows using a narrow coincidence window between the calorimeter and the FICH facilitating low pile-up in the coincidence reconstruction. Its compactness hosting a total of 14 samples as well as the minimal amount of structural material in beam provide excellent conditions for low background and high statistics measurements. The whole experimental set-up was further designed to achieve good performance, especially stability over time as well as effectively tagging the fission events with an efficiency close to 90%.

An experiment aiming at measuring the $^{233}$U $\alpha$-ratio was performed and the results have shown that the developed fission chamber is well suited to tag the prompt fission $\gamma$-rays, hence to have a good control over the fission background in the capture measurement. Results of this measurement will be presented in a separate publication.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

M. Bacak: Software, Validation, Visualization, Investigation, Formal analysis, Writing - original draft, Data curation. M. Aïche: Methodology. G. Bélier: Conceptualization, Investigation, Methodology, Resources, Writing - review & editing. E. Berthoumieux: Software, Validation, Investigation, Conceptualization, Methodology, Project administration, Writing - review & editing, Data curation, Supervision. M. Diakaki: Investigation, Conceptualization, Methodology, Writing - review & editing. E. Dupont: Investigation, Conceptualization, Methodology, Project administration, Writing - review & editing, Funding acquisition, Data curation, Supervision. F. Gunsing: Investigation, Conceptualization, Methodology, Project administration, Writing - review & editing.
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References