New experiment for measuring the delayed neutron yields of ^{238}U fission in the range 1MeV to 19 MeV

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***Abstract*— In nuclear reactors operating in nominal conditions, delayed neutrons (DN) drive the kinetic behavior of the neutron population. Predicting this behavior requires therefore the knowledge of the DN yield per fission, average lifetime, and kinetic of their emission, which is described by precursors groups abundances. Characterizing the uncertainties of these macroscopic data is essential, especially as they are used to design the safety margins of reactors.**

Even for major fissile isotopes such as ^{235}U and ^{239}Pu, discrepancies still appear between the main nuclear data library. In 2018, CEA launched the ALDEN project (Average Lifetime of DElayed Neutrons), gathering several laboratories (CEA/DES and DRF, CNRS/LPSC, LP2i and LPC Caen, ENSICAEN, and University of Caen). This collaboration aims at producing new precise DN macroscopic data for reactor applications.

Three experimental campaigns were conducted in 2018, 2019 and 2021 on the thermal fission of ^{233}U, ^{235}U and ^{239}Pu at Institut Laue Langevin in Grenoble (France), giving high quality results. In February 2023, a new campaign was performed at the PTB Ion Accelerator Facility in Braunschweig (Germany), focusing on the fast fission of ^{238}U. In particular, the DN yield was measured for eight energies between 1.5 and 19 MeV. This paper presents the experimental setup, that was adapted to the new facility and neutron spectrum. The methodology of analysis is also detailed, and some preliminary delayed neutron decay curves are showed.

***Keywords* — Delayed neutrons, ^{3}He proportional counters, uranium 238, fast neutron induced fission**

I. INTRODUCTION

Delayed neutrons (DN) are produced during the beta decay of particular fission products, called “precursors”. Emitted with a mean energy of 500 keV and between 1 ms to 1 minute after the fission, they play a key role in predicting the kinetic behavior of nuclear reactors, even though they represent less than 1 % of the neutrons emitted after a fission . In particular, knowing the average number of DN produced after a fission (called DN multiplicity or yield) and the kinetics parameters (precursors groups abundances) associated to their emission is needed to estimate the effective DN fraction (βeff, fraction of fissions induced by DN), which also defines the limit of criticality accident in nuclear reactors.

For isotopes of main interest for reactor applications, discrepancies still exist between nuclear data libraries (such as JEFF-3.3 and ENDF/B/VIII) [1]. In the case of ^{238}U in particular, very few experimental data are also available, and are usually associated with uncertainties higher than 5% [2]. In 2018, the ALDEN collaboration was therefore settled, gathering laboratories working in the nuclear data field (CEA/DES and DRF, CNRS/LPSC, LP2i and LPC Caen, ENSICAEN and University of Caen). The objective of the collaboration was to produce new experimental DN data with reduced uncertainties, focusing at first on the thermal fission of fissile isotopes.

An experiment was designed, based on an efficient fast neutron detection system. The principle of the measurement was the following: a sample of the investigated isotope is irradiated inside the detector in order to induce fissions and produce precursors. After a certain period of time, the irradiation is stopped and the DN emitted during the precursors decay are counted. Cycles of irradiation/DN counting are repeated to increase the statistics, the duration of both phases being adjusted depending on the DN data that is targeted. The experimental setup was used three times at the PF1b cold neutron beam at ILL (Institut Laue Langevin, Grenoble, France) in September 2018 [1], June 2019 and March 2021 [3]. Very precise measurements (uncertainty lower than 1 % on the DN yield) were produced for the thermal fission of ^{233}U, ^{235}U and ^{239}Pu [4].

After the success of these measurements, it was decided to investigate the fast fission of ^{238}U, and especially the energy dependency of the DN multiplicity. The experimental setup was then adjusted to be used with fast monoenergetic neutrons at the PTB Ion Accelerator Facility (PIAF, Braunschweig, Germany). After being tested on several occasions at the GENESIS facility (Grenoble, France, in February and October 2022) and at PIAF (in December 2022), it was finally used during a two weeks long campaign conducted at PIAF in February 2023), during which eight energies were investigated between 1.5 and 19 MeV.

The present paper focuses on the campaign performed in February 2023 at PIAF. After presenting the experimental setup used, the measurements performed are detailed. The methodology used to analyze the data is then described. Finally, preliminary results are shown and discussed.

II. EXPERIMENTAL SETUP

A. *PTB Ion Accelerator Facility*

The measurements took place in the “Low-scatter hall” of the PTB Ion Accelerator Facility at Braunschweig (Germany). Monoenergetic neutrons flux were produced through the interaction of a solid tritium or gaseous deuterium target and a beam of protons or deuterons delivered by a Tandem accelerator. The solid tritium targets were composed of a titanium layer loaded with tritium and deposited on either a silver or an aluminum backing. The deuterium gas target was 30 mm in length and had a pressure of around 1.6 bar. A molybdenum foil (5 µm in thickness) was placed at its entrance. Table 1 summarizes the characteristics of the targets used during the campaign.

A steering magnet was mounted on the beam line before the experimental hall. It was used to deflect the incident charged particles beam in order to perform the irradiation/decay cycles required for the measurements.

TABLE 1
CHARACTERISTICS OF THE TARGETS USED TO PRODUCE THE INCIDENT MONOENERGETIC NEUTRON FLUX

E_n MeV	Reaction	E_{proj} KeV	Target	Thickness $\mu g/cm^2$	T/Ti mass ratio
1.5	$T(p, n)^3He$	2375	Solid	1831	1.25
2.5	$T(p, n)^3He$	3319	Solid	999	1.18
3	$T(p, n)^3He$	3840	Solid	1831	1.28
4	$D(d, n)^3He$	1740	Gas	-	-
5	$D(d, n)^3He$	2419	Gas	-	-
6.5	$D(d, n)^3He$	3705	Gas	-	-
14.8	$T(d, n)^4He$	215	Solid	1026	1.25
19	$T(d, n)^4He$	2674	Solid	1871	1.25

E_n and E_{proj} are respectively the energies of the neutron produced and of the incident charged particle.

B. *Uranium sample*

The uranium sample was a cylinder of metallic depleted uranium (0.22% in ^{235}U), with 15 mm in diameter, 9.7 mm in height and a mass of 29.5481 g. To ease its manipulation and for radioprotection issues, it was encapsulated in an aluminum container (1 mm in thickness) prior to the campaign (see Fig. 1). A passive gamma counting of the sample was performed in the beginning of the campaign with the HPGe detector of the facility to verify its isotopic composition.

Fig. 1. From left to right: ^{238}U sample before and after encapsulation, scheme of the encapsulation (red: sample, grey: aluminum container, blue: aluminum plug).

C. *Detection system*

The detector LOENIEv2 was already used in the previous ALDEN measurements performed at ILL [1]. It was made of sixteen ^3He proportional counters (PCs) at a pressure of 10 bar, polarized at a high voltage around 2100 V and placed on three concentric rings in a cylinder of high-density polyethylene (HDPE, 46 cm in diameter, 34 cm in length, see Fig. 2). The center of the detector was left free to allow placing the sample under investigation. Delayed neutrons emitted in the sample were thermalized in the HDPE before inducing $^3He(n, t)$ reactions in the PCs. The global geometry of the detector, including the position of each PC, has been optimized in order to ensure an almost flat efficiency (2% relative variation) in the range 0.1 to 1 MeV, the energy range of delayed neutrons emitted inside the sample. Finally, boron rubber sheets covered the inner and outer faces of the HDPE cylinder, absorbing thermal neutrons. These sheets played a key role in reducing the detection of room-returned neutrons as well as limiting the fission of ^{235}U inside the sample from incident neutrons thermalized in the HDPE.

LOENIEv2 was calibrated for delayed neutrons emitted at its center in December 2021 at the NPL facility in England [5]. For the measurements performed at PIAF however, the sample was placed at some distance from the center of the detector (see section D). Therefore, a relative calibration was performed between these two positions with an AmBe source during the campaign.

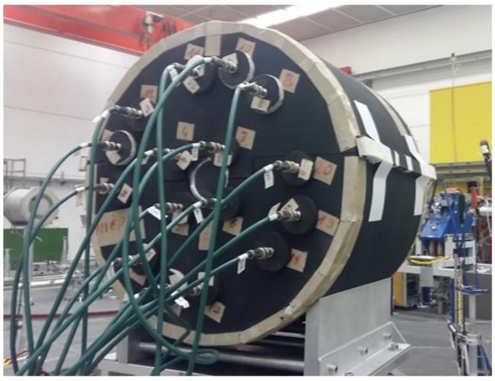

Fig. 2. Picture of LOENIEv2 during the installation. ^{3}He PCs are inserted from the back face of the detector. Inner and outer faces of the PEHD cylinder are covered with boron rubber sheets (in black).

D. Source, sample and detector geometry

To maximize the fission rate inside the sample and be able to count delayed neutrons as soon as the irradiation stops, LOENIEv2 was placed at a few centimeters from the neutron production target, with the sample inside. The detector was aligned thanks to a set of sights, used in association with the lasers and telescopes available in the experimental hall and allowing visualizing the horizontal and vertical axes of the beam line. A specific tool was developed based on a depth gauge to measure the distance separating LOENIEv2 from the target.

The encapsulated sample was placed in a three wings aluminum support ensuring its radial centering. The support was inserted in an aluminum guiding tube placed at the center of the detector. An aluminum stop placed at the end of the tube provided a repeatable and precise positioning of the sample, at around 4 cm from the front face of the detector (see Fig. 3).

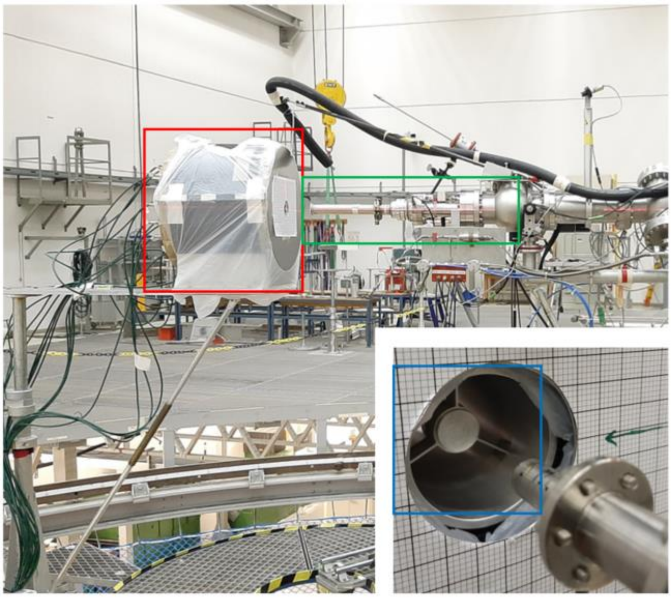

Fig. 3. Placement and alignment of LOENIEv2 (in red) at proximity of the neutron production target (in green). Positioning of the 238U sample inside the detector (in blue).

E. Data Acquisition System

The acquisition system was designed by the ILL instrument control group in 2018 for the first AL33DEN campaign. It was operated using the NOMAD software also developed at ILL [6].

The signals of the sixteen ^{3}He PCs were shaped using a sixteen channels charge sensitive preamplifier card. They were then processed by two synchronized digitizers (CAEN V1724, 8 channels, 14 bits 100 MS/s Flash ADC) running a real time digital pulse processing program (CAEN DPP-HPA). This program allowed detecting the events and obtaining their arrival time and energy (pulses amplitudes). Data were stored in binary files in “list mode” before being further processed. The signal of a neutron monitor placed at 97° and around 2.5 m from the target, already shaped by the facility, was recorded as well using an additional histogram card (recording in histogram mode, no energy determination). Finally, the acquisition system communicated with a monitoring computer through an Ethernet link, allowing transferring a large data rate from the experimental hall to the command room of the facility.

To avoid saturating the system during irradiations (detection of a large amount of incident, prompt and delayed neutrons), the acquisition of the ^{3}He PCs was inhibited during the main part of it, the rising and falling edges left apart. A low frequency generator was therefore used to generate an inhibition signal. It was synchronized with the cycles performed by the steering magnet cutting the neutron production off, thanks to a trigger signal provided by the facility. As the measurements required to record complete cycles of irradiation/decay (especially for group abundances measurements), a synchronized trigger signal was also generated using the same low frequency generator. This allowed launching automatically acquisitions at the beginning of the irradiations.

During irradiations, ^{3}He PCs were subject to a very high reaction rate. Tests have therefore been conducted prior to the campaign in February 2022 at the GENESIS facility in Grenoble (France) to verify that such reactions rates would not affect the efficiency of the counters after the end of the irradiation and that the system was able to recover after the beam stop in less than a few milliseconds.

III. THE EXPERIMENTAL CAMPAIGN OF FEBRUARY 2023

A. Overview of the delayed neutrons measurements

The experimental campaign was conducted over two weeks, during which DN multiplicity was measured for eight energies between 1.5 – 6.5 and 14.8 – 19 MeV by performing short cycles. The durations of these cycles (2/0.2 s, 2.1/0.1 s, 4/0.3 s or 6/1 s) were adjusted depending on the energy due to the variations of the beam cut off duration (see section IV.B). The DN group abundances were also measured for three of these energies (see Table 2) by performing long cycles allowing to count the DN emitted by each group (50/300 s).

For 19 MeV, an additional DN multiplicity measurement was performed with a blank target (same target without tritium) to correct for the neutrons produced by deuteron break-up reactions due to incident deuterons implementing in the target backing.

TABLE 2
LIST OF DN MEASUREMENTS PERFORMED DURING THE CAMPAIGN

Day	Energy MeV	Cycle	Objective
5	14.8	2 / 0.2 s	DN multiplicity
5	14.8	50 / 300 s	Groups abundances
6	4	2.1 / 0.1 s	DN multiplicity
7	5	4 / 0.3 s	DN multiplicity
7 and 8	5	50 / 300 s	Groups abundances
11	2.5	4 / 0.3 s	DN multiplicity
11	2.5	50 /300 s	Groups abundance
12	1.5	4 / 0.3 s	DN multiplicity
12	3	4 / 0.3 s	DN multiplicity
13	6.5	4 / 0.3 s	DN multiplicity
13	6.5	6 / 1 s	DN multiplicity
14	19	6 / 1 s	DN multiplicity

B. Sample fission rate estimation

Two methods were used for each energy to estimate the neutron flux seen by the uranium sample and determine the fission rate inside it during DN measurements. The first one comes from the knowledge that the facility has of the emitted neutron flux. For each energy, two neutron monitors (at 16° and 97 ° and around 2.5 m from the target) were calibrated using a reference long counter. The operation was repeated after placing LOENIEv2 close to the target to account for incident neutrons scattered in the detector. The counting rates inside the neutron monitors were then recorded during each DN measurement, allowing estimating the neutron flux emitted by the source and seen by the sample, thanks to a Monte-Carlo simulation of the experimental setup.

The second method consists in characterizing the neutron flux by dosimetry. Indium, Cobalt, Nickel or Magnesium dosimeters (2 mm in thickness, 15 mm in diameter) were encapsulated in an aluminum container and irradiated during a separate run at the position of the uranium sample. They were then measured by gamma spectrometry either on site using the HPGe detector of the facility (Indium and Magnesium dosimeters mainly) or at the Low-Activity Laboratory at LPSC Grenoble (France) for the radionuclides of high half-life. The fission rate will then be estimated by simulating the irradiation of both the dosimeters and the uranium sample (Monte-Carlo calculations).

A third method was employed for the first energy of the campaign (14.8 MeV). It consisted in activating the not-yet irradiated sample through a thirty minutes long continuous irradiation before estimating the reaction rates of some fission products (^{92}Sr and ^{135}I) by gamma spectrometry. The fission rate was then deduced using the cumulative fission yields of the measured fission products. As this method does not require to simulate the experimental setup, it will be used as a reference to validate the two previously presented methods at 14.8 MeV.

C. Optimization and characterization of the background and beam cut-off

Both the background and beam cut off durations varied significantly with the neutron energy investigated. As these are key points in estimating the DN multiplicity and groups abundances, a consequent amount of time and efforts was dedicated to their optimization and characterization for each energy.

The optimization phase was performed with a dummy container similar to the one encapsulating the uranium sample. The background optimization was conducted by deflecting the incident charged particles beam using the steering magnet (configuration used during the delayed neutrons counting) and acting on the settings of the magnet and the beam line entrance window while monitoring the counting rate inside the ^{3}He PCs. The same procedure was applied for the beam cut off optimization, except this time cycles of irradiation/decay were performed (similar to the ones used for DN multiplicity measurements).

At the end of the optimization phase, the beam cut off was characterized using a dummy container and performing cycles, whereas the background was measured with the uranium sample in place and with the incident charged particles beam deflected.

IV. DATA ANALYSIS STRATEGY AND FIRST RESULTS

A. Methodology of analysis

Each "run" of acquisition produced a binary file containing the data associated to every event recorded by each ^{3}He PCs during the acquisition. Runs performed for DN multiplicity measurement contained several cycles whereas one run corresponded to one cycle for groups abundances measurement. For DN multiplicity measurements, the first runs (around ten minutes) were left out of the analysis, as the population of precursors had not reached its equilibrium yet.

The first step of the analysis process is to convert every binary file into a readable format compatible for instance with the MATLAB environment. Then several steps are taken in order to produce the DN decay curves that will be used to estimate the DN data of interest:

1. PHA spectra are built for each ^{3}He PC, in order to select the energy interval corresponding to neutron detection.
2. Time histograms of the counting rate inside the ^{3}He PCs are generated for each run.
3. Counting rates of each ^{3}He PC is corrected for dead time, by applying a non-extendable model of parameter τ, which was estimated in previous tests performed in February 2022 at the GENESIS facility (Grenoble, France).
4. For each run, the cycles are separated based on a threshold applied to the counting rate summed over all ^{3}He tubes, allowing identifying the beginning and the end of the irradiation.
5. The "DN counting only" time interval is then identified for each cycle (between the end of an irradiation and the beginning of the next one).

6. Finally, the obtained counting rates inside the time interval of interest are summed over all ^{3}He PCs and all cycles.

During these steps, the actual duration of the cycles performed are also estimated.

The next steps consists in estimating the background observed during the DN counting, the fission rate inside the sample during irradiations (using the three methods introduced in section 0) and the efficiency of LOENIEv2 towards DN emitted inside the sample (based on the relative calibration performed with an AmBe source). Once these steps are completed, the DN multiplicity can be deduced using (1):

$$c(t) = b_{off} + \nu_d F_0 \sum_k a_k \varepsilon_{d,k} \frac{(1-e^{-\lambda_k t_{irr}})e^{-\lambda_k t}}{1-e^{-\lambda_k t_m}}, \qquad (1)$$

Where *c(t)* is the counting rate at *t* ($t = 0$ corresponding to the end of irradiation), b_{off} the background during DN counting, ν_d the DN multiplicity, F_0 the fission rate inside the sample, a_k the abundance of group k, $\varepsilon_{d,k}$ the efficiency of LOENIEv2 for DN of group k, λ_k the decay constant associated to group k, t_{irr} the duration of the irradiation and t_m the duration of the irradiation/decay cycle.

B. Preliminary results: DN decay curves, signal to noise ratio and veto time

Fig. 4 presents two example of DN decay curves obtained for DN multiplicity measurements at 14.8 and 2.5 MeV. DN counting rates varied a lot between energies, as could be expected due to the variations of fission cross section, DN multiplicity and flux emitted by the targets. The measured background also showed some variations with the neutron energy, leading to signal over noise ratios ranging from 12 to 163 (see Table 3).

Substantial variations were also observed during the campaign concerning the beam cut off duration, depending on the energy investigated, as illustrated in Fig. 4. During this period of time, not only delayed neutrons are detected, but also remaining incident neutrons. Therefore, a veto time has to be applied after the identified end of irradiation, during which the neutron detections are not taken in consideration for the estimation of the DN multiplicity. Although it is most likely insignificant compared to the whole irradiation time, the impact of the additional fissions that are induced during this time on the model described by (1), which considers perfectly squared irradiations, will have to be investigated.

Table 4 presents the preliminary fission rates estimated using the methods introduced in section III.B, for 14.8 MeV. A good agreement is observed between the results given by the activation of the ^{238}U sample and the flux calibration using dosimeters with high-energy threshold reactions. The method based on the PIAF neutron monitors calibration and monitoring to determine the neutron flux however leads to an apparent underestimation of the fission rate. More analysis is required to understand where this discrepancy comes from.

TABLE 3
PRELIMINARY RESULTS OBTAINED FOR EACH ENERGY: COUNTING RATE AFTER THE VETO TIME FOLLOWING THE END OF THE IRRADIATION (INCLUDING DN AND BACKGROUND, BKG), BACKGROUND MEASURED WITH THE SAMPLE IN PLACE AND THE BEAM DEFLECTED, SIGNAL TO NOISE RATIO OBTAINED AND VETO TIME ESTIMATED.

Energy MeV	DN + bkg counting rate (s^{-1})	Bkg counting rate (s^{-1})	Signal to noise ratio	Veto time ms
1.5	54	0.7	76	70
2.5	76	0.7	108	70
3	134	9.3	13	70
4	101	0.8	125	60
5	148	0.9	163	73
6.5	184	4.8	37	100
14.8	450	16	27	15
19	38	2.9	12	80

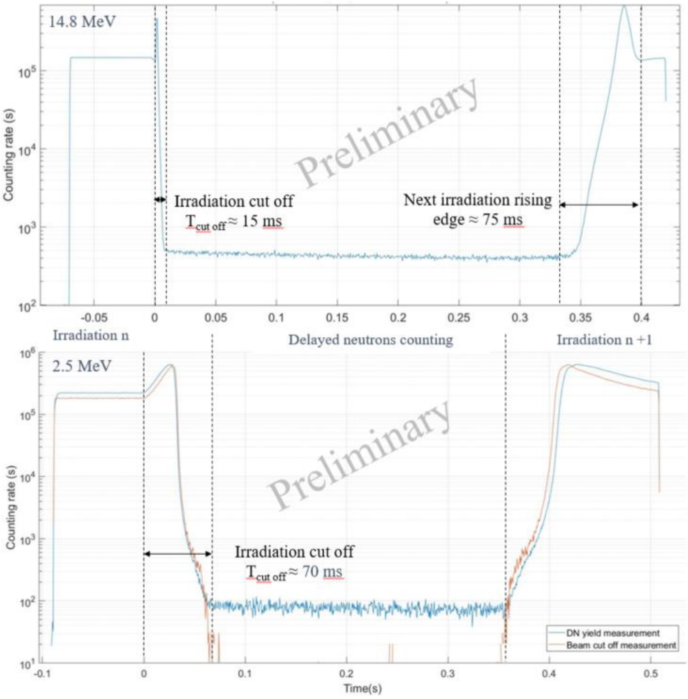

Fig. 4. Decay curves obtained for 14.8 (up) and 2.5 MeV (bottom). Irradiation cut off starts at $t = 0$. Inhibition of the ^{3}He PCs acquisition stops approximately 0.1 s before the beginning of the cut off (counting rate increasing from 0 to more than 10^5 counts/s. For 2.5 MeV, the counting rate obtained during the beam cut off characterization with a dummy container is also displayed in red.

TABLE 4
PRELIMINARY ESTIMATIONS OF FISSION RATE INSIDE THE SAMPLE DURING IRRADIATIONS AT 14.8 MEV USING THE THREE METHODS PRESENTED IN SECTION III.B

Method	Reaction (Threshold, MeV)		Fission rate (x 10^5 fissions/s)
Flux monitoring (PIAF)	-		1.784 (3.5 %)
Dosimetry	^{115}In (n, n') ^{115m}In	(≈0.340)	1.408 (1.2 %)
	^{59}Co (n, p) ^{59}Fe	(≈3)	2.016 (3.0 %)
	^{60}Ni (n, p) ^{60}Co	(≈4)	2.195 (10.1 %)
	^{24}Mg (n, p) ^{24}Na	(≈6)	1.920 (1.1 %)
	^{115}In (n, 2n) ^{114m}In	(≈9)	1.969 (3.2 %)
	^{59}Co (n, 2n) ^{58}Co	(≈11)	2.072 (3.1 %)
^{238}U sample activation	-		2.026 (2.3%)

V. CONCLUSIONS

Since 2018, CEA has conducted new measurements of DN macroscopic data such as multiplicity and groups abundances, in the framework of the ALDEN collaboration. An experimental setup was designed to count the DN emitted from an irradiated sample placed at the center of an efficient neutron detector. This setup has been used three times over the last years at ILL on the cold neutron beam PF1b, with fissile samples (^{233}U, ^{235}U and ^{239}Pu).

Started in 2021, a new project inside the collaboration focused on measuring the DN data for the fast fission of fertile isotopes. The setup was adapted to be used at the PTB Ion Accelerator Facility in Germany. Two weeks of measurements were performed in February 2023, during which DN data associated to the fast fission of ^{238}U were measured for eight incident neutron energies between 1.5 and 19 MeV.

Overall and despite the change of installation and incident neutron spectrum, the campaign was a success, as data were acquired for all energies planned. The analysis is still in progress, but preliminary results suggest that the measurements performed were of good quality.

Finally, a second campaign is planned in September 2023, focusing on energies between 7 and 14 MeV. It will benefit from the feedback of the measurements conducted in February, including for example the need to better characterize the beam cut off profile at the end of each irradiation.

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