



Ricerca di Sistema elettrico

# Dati Nucleari per la Sicurezza Reattore – PAR 2016: Messa a punto di una metodologia di valutazione di sezioni d'urto per analisi margini di sicurezza: tecniche e campagna sperimentale per nuove valutazioni delle sezioni d'urto di cattura neutronica degli isotopi dispari del Gadolinio

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DATI NUCLEARI PER LA SICUREZZA REATTORE – PAR 2016: MESSA A PUNTO DI UNA METODOLOGIA DI VALUTAZIONE DI SEZIONI D'URTO PER ANALISI MARGINI DI SICUREZZA: TECNICHE E CAMPAGNA SPERIMENTALE PER NUOVE VALUTAZIONI DELLE SEZIONI D'URTO DI CATTURA NEUTRONICA DEGLI ISOTOPI DISPARI DEL GADOLINIO

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**Titolo**

**Implementation of a cross section evaluation methodology for safety margins analysis: techniques and experimental campaign for new evaluations of neutron capture cross sections of Gadolinium odd isotopes**

**Descrittori**

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**Sommario**

The second and third generation Light Water Reactors (LWRs) are often equipped with fuel with gadolinium-enriched pins with the aim of extending the in-pile fuel life cycle without introducing any new safety issues at the Beginning Of Life (BOL). Gadolinium odd isotopes cross sections are crucial in assessing the neutron budget which constraints the economic performance and the safety features of a reactor core.

This technical report follows the previous one on the justification for a new evaluation of the neutron capture cross-sections of <sup>155</sup>Gd and <sup>157</sup>Gd using new experimental data from measurements carried out at the European Research Council Center (CERN) in the framework of the n\_TOF Collaboration to which ENEA participates as official member.

The first part of the report describes the CERN neutron time-of-flight facility n\_TOF, its unique features, and the methodologies, including the detection techniques adopted for the measurements of neutron-capture cross sections of Gd odd isotopes.

The second part presents the experimental campaign at n\_TOF, performed from 16th June to 8th July 2016, which lasted for about 21 days.

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## 1. Introduction

“Burnable poisons” are very important in the neutronic balance and the safety features of Light Water Reactors (such as PWRs, BWRs, or VVERs) of 2nd and 3rd Generation, because they play a crucial role for their very high thermal neutron-capture cross sections, higher than the thermal neutron-fission cross section of  $^{235}\text{U}$ .

Gadolinium isotopes are important because they allow to increase the fissile material percentage in fuel rods without any impact on the reactor safety. In fact, as soon as the fuel in the FAs is burnt during the operation of a given reactor, both  $^{235}\text{U}$  and burnable poisons are depleted so that the compensating effect of the poisons is neutralized at a point in the cycle of the fuel at which the remaining amount of fissile material can be controlled easily and safely by other available means.

Gd odd isotopes are also used to better estimate the s.c. “residual reactivity penalty” which is fundamental for evaluating the time a given FA can be used at full power. Accurate predictions of the burning rate of odd-Gd isotopes are important in the forecast of the appearance of the FA reactivity peak and its intensity. In turn, these two parameters are of utmost importance in the assessment of the criticality safety margins for the storage of partially burnt fuel inside Spent Fuel Pools (SFPs) of reactors, especially during postulated loss-of-coolant or loss-of-cooling accidents at these storage facilities [1].

Moreover for CANDU reactors Gd-odd isotopes are used to reduce or eliminate risks due to criticality in the form of nitrate mixed in the heavy water moderator [1].

The necessity of a complete review of the Gadolinium-155 and -157 isotope cross sections was fully illustrated in previous works of the same working group [1], [2].

Against this background, ENEA decided to propose new precise and accurate measurements of Gadolinium neutron-capture cross sections in the framework of the CERN n\_TOF Collaboration, in which ENEA participates as official member.

In 2015, a formal Proposal was submitted by ENEA and INFN to the CERN ISOLDE and n\_TOF Committee with the aim of performing neutron-capture cross section measurements of  $^{155}\text{Gd}$  and  $^{157}\text{Gd}$  isotopes between thermal energy and 1 MeV. The proposal was accepted, and it was allocated a dedicated beam time at the CERN n\_TOF facility, for the experiment to be performed in the Summer 2016.

Several research institutions express interest in this activity: UniBO, CEA, JRC-Geel and PSI [3], while statements of interest arrived also from IRSN, ORNL and Brookhaven National Lab (S. Mughabgab) .

After the approval of the CERN Scientific Committee, 4 high-purity Gd samples in metallic form with a cross contamination of the two isotopes of less than 1% were acquired by Oak Ridge National Laboratories early in 2016. All samples had the same dimension, and were prepared as discs of 1 cm of radius. The measurements were performed between June and July 2016 in the first experimental area (EAR1) of the CERN n\_TOF facility, using the Time-of-Flight (TOF) technique and were carried out with an array of four fast hydrogen-free liquid scintillator



detectors specifically designed for the detection of gamma rays produced by the de-excitation of the compound nucleus formed just after the capture event.

Preliminary studies, carried out before the submission of the proposal, showed that the cross section for both isotopes drops by several orders of magnitude going above 0.1 eV. As a consequence, a unique sample for each odd isotope could not be used to perform the measurement in the whole energy range of interest. In particular to avoid problems related to sample self-shielding, very thin samples needed to be used in order to measure the cross sections near thermal energies, while above 1 eV thicker samples had to be used in order to obtain a good signal-to-background ratio, and to collect the necessary statistics in a reasonable time [3].

In addition to the four Gadolinium samples, ancillary measurements had to be performed for different purposes: normalization to absolute cross section values and background estimation. Therefore during the experimental campaign, a Gold sample ( $^{197}\text{Au}$ ) was used for normalization purposes, while a Graphite and a Lead samples were used to study the different type of background in EAR1. Moreover, black resonance filters (Ag, Co, W, Cd) positioned along the flight path were used to determine the energy dependence of the background; they were chosen thick enough that the neutron beam was completely absorbed at the energies of the largest resonances.

The details of the experimental campaign and the preliminary results of Gd-odd isotopes are presented in the next sections of the present document.

## 2. The nTOF facility

The CERN time-of-flight facility [4,5,6] was proposed by Carlo Rubbia and collaborators and became operative in 2001. The n\_TOF facility supplies a pulsed beam of neutrons which are produced by spallation of 20 GeV/c protons impinging on a massive lead target. The intense proton beam, with  $7 \times 10^{12}$  protons per bunch on average delivered by the CERN Protosynchrotron, makes this facility most suited for the measurement of energy-dependent cross-sections, thanks to the high instantaneous neutron flux. The neutron-producing target is equipped with a moderating system so to generate a neutron spectrum ranging on eleven order of magnitudes: from thermal to about 1 GeV. In addition the 185-m flight path results in an excellent energy resolution ( $\Delta E/E \approx 10^{-4}$  at 1 keV). All these features are well-suited to perform high-accuracy and high-precision measurements of neutron-induced reaction cross sections and make n\_TOF a unique tool for nuclear data measurements.

More in details, the spallation target is composed by 1.3 tons of lead in cylindrical form (40 cm in length, 60 cm in diameter), surrounded by 1 cm of water, and a subsequent layer of 4 cm of borated water, separated by a thin aluminium window. The neutron beam produced by protons impinging on the lead target is cooled by a water layer which surrounds the target itself, while the layer of borated water acts as a moderator, so to produce an "isoletargic" spectrum, i.e. with uniform distribution in lethargy units. Spallation is the most prolific neutron source: in fact, at the proton energy of 20 GeV/c about 300-400 neutrons per proton can be generated

by this mechanism. Neutrons emerging from the spallation target are horizontally canalized along a long flight-path in a vacuum tube for about 185 meters up to the first experimental hall (EAR1).

Along the particle transport line a system of two collimators are used to narrow and shape the beam, a magnet bends charged particles in order to remove them from the beam, and a series of iron and concrete shields are used to reduce the background produced in the spallation process. In fig. 1 the layout the CERN accelerator complex together with a schematic view of the n\_TOF facility are presented.

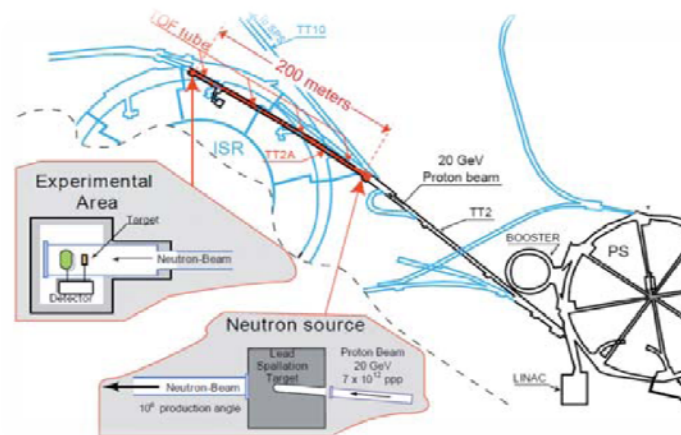
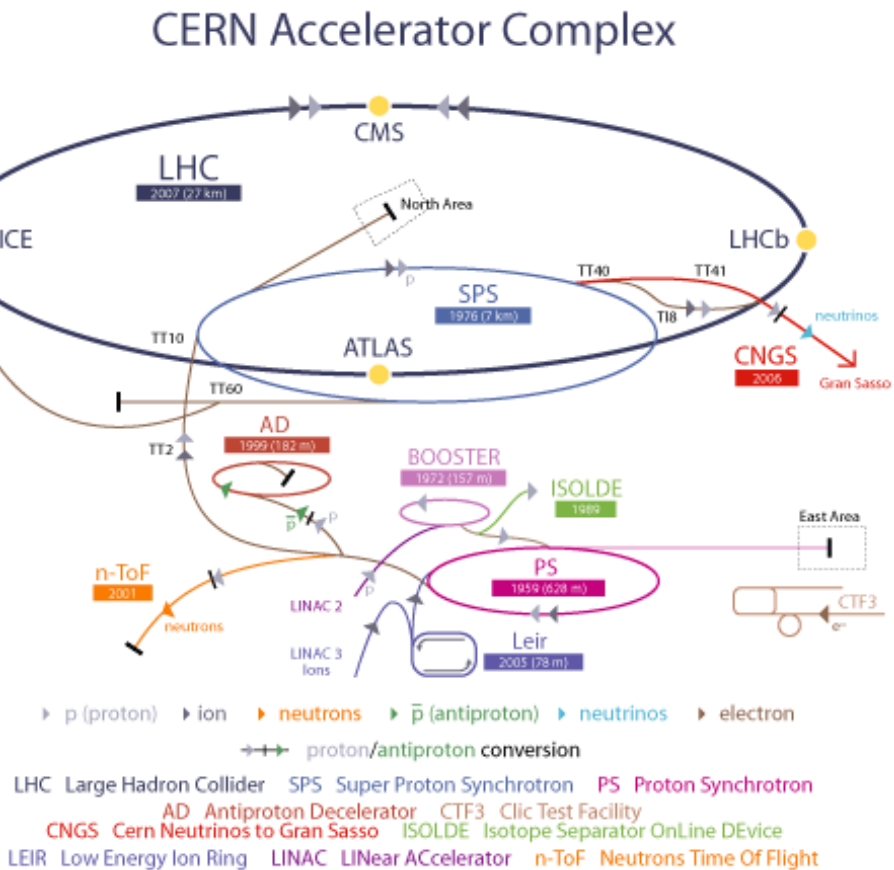


Fig. 1 Layout of CERN complex and schematic map of the n\_TOF facility at CERN [6].

Another important feature of the n\_TOF neutron beam derives from the low repetition rate of the primary proton beam coming from the CERN Protosynchrotron. The low duty cycle, of about 1 Hz, prevents the overlapping of low-energy neutrons from one bunch with the fastest neutrons from the following bunch.

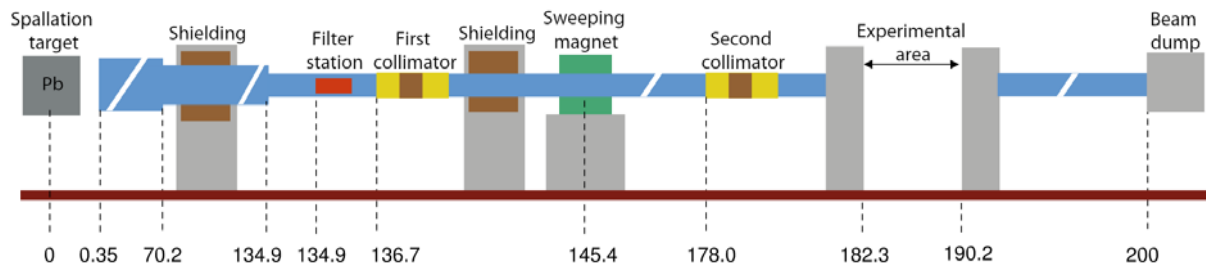


Fig.2 Sketch of the n\_TOF neutron beam line for the EAR 1 from the spallation target to the beam dump. [5]

The very-high intensity of the n\_TOF neutron source of  $2 \times 10^{15}$  neutrons/pulse produced after the spallation target reduces to  $1.5 \times 10^7$  neutrons/pulse at the sample position, where the neutron beam has a diameter of 2 cm (see fig.2).

Since 2001 EAR1 was the only experimental area at n\_TOF. In 2014 a second shorter beam line (EAR2) came into operation. The distance of the new experimental area, which is located perpendicular to the flight path towards EAR1, is shorter by a factor of 10, compared to EAR1. Consequently, the flux in EAR2 is almost two orders of magnitude higher (a factor of between 30 and 100, depending on the energy) at the expense of a lower energy resolution. The high neutron flux in EAR2 allows to expand the experimental program, with measurements on radionuclides with half lives as short as few tens of days, never measured at other neutron time-of-flight facilities. In fact, the combination of the higher flux and the shorter time interval results in an increase of the signal-to-noise ratio of a factor of 250 which is not only useful for cross section measurements of radioactive samples, but also of samples having very low cross sections, and available in small quantities.

The excellent features of the n\_TOF facility are coupled with an innovative data-acquisition system based on flash ADCs with 14 bit resolution and 1 GHz sampling rate and with cutting-edge detectors especially designed for n\_TOF.

Gadolinium odd isotopes neutron capture cross section measurements take advantage of the first beam line (EAR1) and were carried out by an array of detectors fulfilling special requirements. The detectors and the measurement technique are fully described in the next paragraphs.

### 3. The time-of-flight method

In order to resolve the resonant structure of the neutron cross-section which spans over a wide energy range (from thermal to MeV), high resolution measurements have to be performed.



For facilities like n\_TOF, in which a pulsed neutron beam covers a well-known distance to reach the sample to be studied, the time-of-flight (TOF) technique is employed. This method relies on the measurement of the time needed for a neutron to reach the sample at the measuring station. This quantity is used to determine the kinetic energy of neutrons travelling along the beam line.

The kinetic energy  $E_n$  of neutron with speed  $v = \frac{L}{TOF}$  can be expressed as:

$$E_n = E - mc^2 = \sqrt{c^2 p^2 + m^2 c^4} - mc^2 = mc^2 (\gamma - 1)$$

where  $\gamma = (1 - v^2/c^2)^{-1/2}$  is the Lorentz coefficient, and  $c$  is the speed of light. The first term of the series expansion gives the classical expression for the neutron kinetic energy, which can be used below a few keV:

$$E_n = \frac{1}{2} m v^2 = \alpha^2 \frac{L^2}{t^2}$$

Where  $\alpha = 72.29 \sqrt{\frac{eV}{m}} \mu s$ , when the flight path  $L$  is measured in meters, the kinetic energy in eV and the time in microseconds.

The relative energy resolution of a TOF facility is:

$$\frac{\Delta E_n}{E_n} = 2 \cdot \sqrt{\left(\frac{\Delta t}{t}\right)^2 + \left(\frac{\Delta L}{L}\right)^2}$$

Therefore, as already pointed out, the best energy resolution can be gained by using long flight paths and by producing neutrons within the shortest time interval [5]. It is worth noticing that the time spread of the primary proton bunch is only 7 ns long.

#### 4. Methodology and measurements

In capture measurements, cross sections are determined by the detection of the prompt gamma cascade, emitted in the de-excitation of the compound nucleus which is created during the capture process.

The experimental observable is the capture yield ( $Y$ ), which represents the fraction of the incident neutron which undergoes a capture reaction in the sample (and generating a signal in the detection system).

The experimental yield  $Y_x^{meas}(E_n)$  can be calculated from measurable quantities as:

$$Y_x^{meas}(E_n) = \frac{C(E_n) - B(E_n)}{\varepsilon_x(E_n) \cdot \varphi_n(E_n)}$$

where  $C(E_n)$  and  $B(E_n)$  are the total and background counts per neutron pulse respectively,  $\varepsilon_x$  is the detection efficiency for the reaction of interest, and the  $\varphi_n(E_n)$  is the flux impinging on the sample under investigation. This last quantity needs to be evaluated with the same accuracy as for the capture counts.

However, since in most cases  $\varepsilon_x = f(A, \Omega, P)$  depends from:

- A the effective sample area intercepted by the beam;
- $\Omega$  which is the solid angle subtended by the detector;
- P the interaction probability in the sample for a given energy;

an evaluation of the  $Y_x^{meas}(E_n)$  can then be a very difficult task.

For that reason, it is possible to define a normalization factor N which groups together all these parameters which are independent of the neutron energy. In this case the normalization can be determined using the so-called saturated resonance method.

Making that assumption, the previous formula becomes:

$$Y_x^{meas}(E_n) = N \frac{C(E_n) - B(E_n)}{\varphi_n(E_n)}$$

The corresponding calculated quantity  $Y_x^{th}(E_n)$  can be related to the capture cross section as follows:

$$Y_x^{th}(E_n) = (1 - e^{-n\sigma_t(E_n)}) \frac{\sigma_x(E_n)}{\sigma_t(E_n)}$$

where n is the areal density of the sample in atom/barn and  $\sigma_t$  and  $\sigma_x$  are the total and reaction cross sections expressed in barn, respectively. If the capture cross section is high and the sample has adequate thickness, the measured yield is equal to 1. In that way it is possible to determine N.

On the other hand, when the sample is extremely thin it is possible to simplify the expression as follows:

$$Y_x^{th}(E_n) \approx n\sigma_x$$

If the measurement is performed on a neutron standard cross section such as  ${}^6\text{Li}$  or  ${}^{10}\text{B}$ , whose (n,a) cross section is considered as a reference, the previous relations can be used to extract the neutron flux [5]:

$$\varphi_n(E_n) = \frac{C(E_n) - B(E_n)}{\varepsilon_x \cdot (E_n)n \cdot \sigma_x}$$

In order to perform an evaluation of the flux with the required precision and accuracy, during the experimental campaign several detectors based on different conversion reactions were used.

Combining their results in a consistent way, by several dedicated measurements during different experiments and campaigns, in different energy ranges, it was possible to determine the flux from thermal to 200 MeV with rather small systematic and statistical error.

Once known both the normalization factor N and the flux  $\varphi_n(E_n)$ , it is possible to determine the cross section of interest.

## 5. In-Beam gamma ray-background

The spallation mechanism and the different interactions of the produced neutrons with the structures of the facility, caused a sizable production of gamma-rays and charged particles, which can reach the experimental halls [5].

The gamma-ray background consists of a prompt component (TOF > 1  $\mu\text{s}$ ) and a delayed one (TOF > 1  $\mu\text{s}$ ) mostly due to capture events from the cooling and moderator system, as well as from structural material around the spallation target.

The prompt component of the gamma-ray background is commonly named gamma-flash, and is accompanied by a huge amount of charged particles which are produced in the target and is typically used to synchronize in time the detectors in the experimental area.

These components affect the detectors used at n\_TOF in different manners, depending on the detector type and its location inside the experimental halls. Generally, gamma-flashes blind the detectors for times of magnitude of microseconds, so preventing the detection of the neutrons with highest velocities which fly along the line.

In order to minimize the effects of the so-called gamma flash on the detectors used for the capture measurements and its neutron sensitivity, a novel type detector (C6D6) with very special features was developed by the INFN Legnaro Group [8].

## 6. Gamma capture measurement with C6D6 detectors


The n\_TOF Collaboration has developed over the years a series of highly-featured detectors suited for different type of measurements and fulfilling cogent requirements. Among them, in order to study the resonance structure of neutron induced capture cross-sections of different reactions, two different detection systems are currently being used: a first one based on H-free deuterated benzene liquid scintillators (C6D6), and a second one is a  $4\pi$  detector based on BaF<sub>2</sub> crystals (Total Absorption Calorimeter -TAC) [7]. Both systems are used in combination with different techniques which are suitable for measurements on samples characterized by different properties.

H-free deuterated scintillators, developed for nTOF by the Legnaro - INFN Group, allows to reveal the gamma rays following neutron captures, with low sensitivity to sample-scattered neutrons.

In fact, C6D6 detectors (see fig. 3) are detectors based on liquid scintillators encapsulated in a carbon fiber cell, coupled with a small photomultiplier tube (PMT) with a very thin window. This setup has the disadvantage of a worse light collection. In order to minimize this problem, the active cell has a cylindrical part with a 45° conical shape to improve light collection at the photocathode. This conical part works both as light guide and as active scintillation volume. [8]



Fig. 3. Image and sketch of a C6D6 detector: in evidence, the main module, and the expansion system which prevents the formation of air bubbles inside the detector head and compensates thermal expansion [8].

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Such a system satisfies the following mandatory requirements:

- The detection efficiency for a capture event is independent of the subsequent gamma-ray cascade, i.e. independent of the multiplicity of the gamma-ray spectrum and the gamma-ray distribution;
- The sensitivity to scattered neutron is extremely low;
- The detector has a very good timing response, with very fast recovering time from the so-called gamma flash created in the spallation process.

Because C6D6 detectors are organic scintillators, they are very fast (only few nanoseconds for the light decay time) so showing a recovery time after the gamma-flash, that is very small compared to other types of detectors, so allowing measurements on neutrons with higher energies.

The disadvantage is linked to the low efficiency of these detectors, which requires special techniques of analysis (the so-called "Pulse Height Weighting Technique").

The second system is instead a total absorption calorimeter (TAC), that is, a  $4\pi$  apparatus that allows to fully record the entire de-excitation cascade of the compound nucleus. Its main drawback is the relatively high neutron sensitivity and for this reason was not used for the Gd cross section measurements.

## 7. Total Energy Detection Technique

The Total Energy Detection Technique requires that the efficiency of the detection system is directly proportional to the total radiative energy emitted by the decay of the compound nucleus formed during the capture process.

In order to comply with this requirement, detectors must have efficiency low enough to achieve the proportionality between the gamma-ray detection efficiency to the gamma-ray energy [9], [10].

Such a proportionality can be achieved with the so-called Pulse-Height Weighting Technique (PHWT). The goal is achieved artificially modifying the detection efficiency so to make it independent of both the cascade path and the energy of a particular gamma-ray being detected. In other words, it is always possible to find a function  $W(E)$  of the pulse height such that its convolution with the pulse height distribution of the capture spectrum becomes proportional to the total energy released.

Systems fulfilling these requirements are based on the use of detectors characterized by extremely low gamma-ray efficiency that is assumed to be proportional to the gamma-ray energy:

$$\varepsilon_{\gamma} = k \cdot E_{\gamma}$$

When the gamma-ray detection efficiency is very small ( $\varepsilon_{\gamma} \ll 1$ ) so that essentially only one gamma-ray out of the capture cascade is registered at a time, the detection efficiency in capturing events can be approximated by:



$$\varepsilon_c = 1 - \prod_i (1 - \varepsilon_{\gamma,i}) \approx \sum_i \varepsilon_{\gamma,i}$$

Under these conditions the detector efficiency for a single event  $\varepsilon_c$  is directly proportional to the total released energy  $E^*$  by the compound nucleus formed during each capture event, and independent of the cascade path:

$$\varepsilon_c \approx k \sum_i E_{\gamma,i} \approx kE^* = k(B_n + E_{n,cm})$$

where  $B_n$  and  $E_{n,cm}$  are the neutron binding energy and the neutron kinetic energy in the centre of mass system respectively.

Therefore, the detection efficiency has been made independent of the cascade path and equal to the known excitation energy of the compound nucleus.

## 8. Pulse Height Weighting Technique (PHWT)

In order to comply with the requirements of the previous paragraph, some mathematical manipulation of the detector response is needed.

The measured counts for each deposited energy must be weighed with an energy (pulse height) dependent weighting function (WF) which needs that the response distribution of the detector is very well known.

A weighting function  $W(E_d)$  is defined such that it follows the relationship:

$$\int_0^{\infty} R_d(E_d, E_{\gamma}) W(E_d) dE_d = kE_{\gamma}$$

where  $R_d(E_d, E_{\gamma})$  is the detector response, i.e. the probability that a gamma ray with an energy  $E_{\gamma}$  results in a deposited energy  $E_d$ . The overall efficiency for detecting a gamma-ray of energy  $E_{\gamma}$ , corresponds to:

$$\int_0^{\infty} R_d(E_d, E_{\gamma}) dE_d = \varepsilon(E_{\gamma})$$

Using the weighting functions, a weighted spectrum can be obtained as follows:

$$C_w(T_n) = \int C(T_n, E_d) W(E_d) dE_d$$

where  $C(T_n, E_d)$  is the distribution of the energy  $E_d$  deposited by the gamma-ray detected at the time-of-flight  $T_n$ , related to the kinetic energy  $T_n$  of the incident neutron.

In order to calculate  $R_d(E_d, E_\gamma)$  Monte Carlo codes (GEANT and MCNP) are routinely used.

The weighting function is obtained by expressing  $W(E_d)$  as a smooth function of the observed deposited energy  $E_d$ , whose parameters are obtained by minimizing for a number of gamma-ray responses, the following expression:

$$\chi^2 = \sum_j \left( kE_{\gamma,j} - \int_{E_L}^{\infty} R_E(E_d, E_{\gamma,j}) W(E_d) dE_d \right)^2$$

## 9. Neutron flux

In cross section measurements it is a key issue to know the total number of neutrons impinging on the target as a function of the neutron energy. In fact the precise and accurate knowledge of the flux over the entire energy range of interest for the measurement to be done (i.e. the number of neutrons per incident proton pulse integrated over the full spatial beam profile) is crucial for the data reduction and analysis. In addition, the accuracy of the absolute value and the energy distribution with which the neutron flux is known has a deep impact on the accuracy achievable in the cross section measurements.

During the Gadolinium experimental campaign, the flux was determined combining the measurements of several detectors based on different conversion reactions:  ${}^6\text{Li}(n,\alpha)t$ ,  ${}^{10}\text{B}(n,\alpha){}^7\text{Li}$ ,  ${}^{235}\text{U}(n,f)$ . The cross sections of these reactions are considered as a neutron standard and therefore they can be considered as a reference in a well defined energy range.

The results of these measurements are used for the determination of the so-called evaluated neutron flux [12], which in turn will be used for the determination for the Gd-odd isotopes neutron-capture cross section, as explained above.

The following detectors were used for performing these measurements:

- i) a solid-state detector equipped with  ${}^6\text{Li}$  converter;
- ii) a MICRO-MEGAS detector equipped with  ${}^{10}\text{B}$  and  ${}^{235}\text{U}$  converters;
- iii) a calibration fission chamber equipped with  ${}^{235}\text{U}$ ;
- iv) a set of Parallel Plate Avalanche Counter (PPAC) with  ${}^{235}\text{U}$ .

The combination of the measurements performed with these detectors has allowed to cover a wide energy range up to 250 MeV. For a fine characterization of the dips due to neutron absorption in the Al layers present in the beam line, a dedicated MC simulation has been performed. It also permitted to extend the evaluation of the  $n_{\text{-TOF}}$  flux up to the GeV region [11].

## 10. Detector calibration and stability of the detection system

Energy calibration for the 4 C6D6 detectors was performed by standard radiation sources:  $^{137}\text{Cs}$ ,  $^{88}\text{Y}$ , and composite Americium-Berillium and Curium-Carbon gamma-ray sources whose characteristics are showed in the following table:

<i>Isotope</i>	$E_{\gamma}[\text{KeV}]$	$E_{max}^C [\text{KeV}]$
$^{137}\text{Cs}$	661.7	477.3
$^{88}\text{Y}$	898	699.1
	1836	1611.8
<i>AmBe</i>	4.4E+3	4.16E+3
<i>CmC</i>	6130	5885

Table 1. Main features of the calibration sources used during the Gd campaign.

For each detector, these peaks were used to link the channel number from the fADC to the corresponding Compton edge.

The use of calibration sources is also important to check the stability of the detector during the measurement campaign. In fact, comparing different spectra acquired regularly every 3 days for the same calibration sources it is possible to have evidence of changes in the gains, which can be adjusted using different calibration curves.

As there are three key interaction mechanisms of gamma radiation with matter:


- Photoelectric effect;
- Pair production
- Compton scattering;

for the energy of typical gamma-ray sources, the Compton edge is the most probable one, also considering the reduced volume of the C6D6 detectors.

From kinematic considerations, the maximum energy transferred by the gamma ray (Compton edge) is given by the following formula:

$$E_{max}^C = \frac{2E^2}{m_E c^2 + 2E}$$

Due to the gaussian energy resolution of the C6D6 detectors, Compton edge does not show a sharp slope, but it also shows a gaussian shape.

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C6D6 calibration was performed using a fit of the Compton edge and identifying the Full Width at Half maximum (FWHM).

Each FWHM trend versus energy was then fitted with a linear function in order to obtain the calibration line.

## 11. Samples

Cross section measurements require samples as isotopically pure as possible in order to avoid any biases attributable to contamination (i.e. isotopes different from those of interest).

The samples used in the experimental campaign for the new evaluation of neutron capture cross section of Gd-odd isotopes were acquired from Oak Ridge National Lab (ORNL). All the samples are circular in shape with a radius of 1 cm in order to cover the same fraction of the neutron beam with the aim of minimizing the effect of the so-called Beam Interception Factor (BIF<sup>1</sup>) on the measurements.

Samples were shipped in an airtight underpressurized cask (to avoid oxidation), so at the beginning of the experiment they were extracted from it with the maximum care in order to avoid damages because they are extremely fragile and need to be handled with maximum care. After weighing them, and in order to prevent oxidation, samples were sandwiched between two Mylar foils and centered in an annular frame that allows the correct positioning along the line during irradiation. The Mylar foils were stretched with a specific tool and then glued to the frames. At the center of each frame the samples were positioned using a very small drop of glue<sup>2</sup>.

Samples were centered using a jig and a hollow metallic cylinder aligned with the annular frame. A cylinder made of Teflon was used to drive the samples in the hole, so ensuring the correct positioning at the center of the annular frame.

Moreover, in order to verify that the drops do not interfere with the measurements to be done, evaluations of that impact were done performing measurements with frames in absence of Gd samples but using the same quantities of glue needed to arrange each sample in the center of each frame. These evaluations based on specific measurements performed in EAR1 demonstrate that the glue drops have no impact on the measurements to be done.

The following figures show the operations just described.

<sup>1</sup> The so-called Beam Interception Factor (BIF) is the fraction of the neutron beam intercepted by the sample aligned along the flight path. The choice of using samples having the same geometry and dimensions complies with the need of avoiding further normalizations which are of particular importance when the area of the sample being measured is smaller than the neutron beam profile.

<sup>2</sup> Epossidic bi-component.

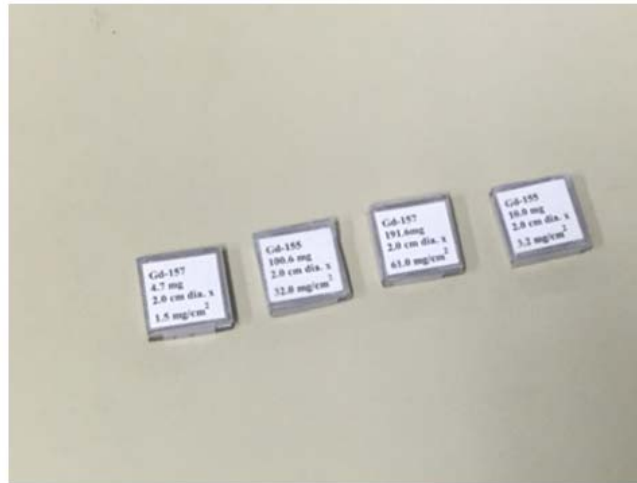
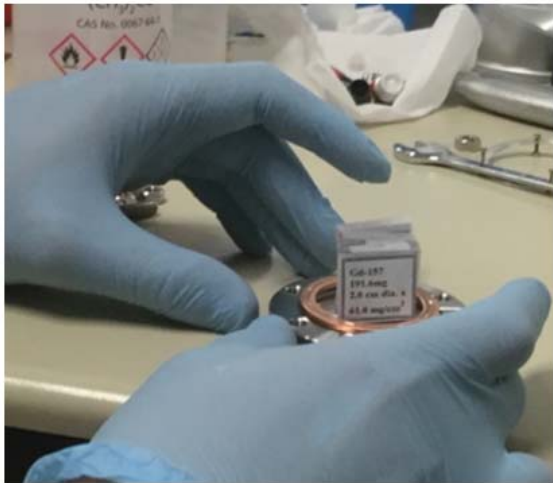


Fig. 4 Gadolinium-odd samples as shipped in an airtight underpressurized cask (upper left hand corner). On the bottom right one: the four Gd samples used for the experimental campaign.



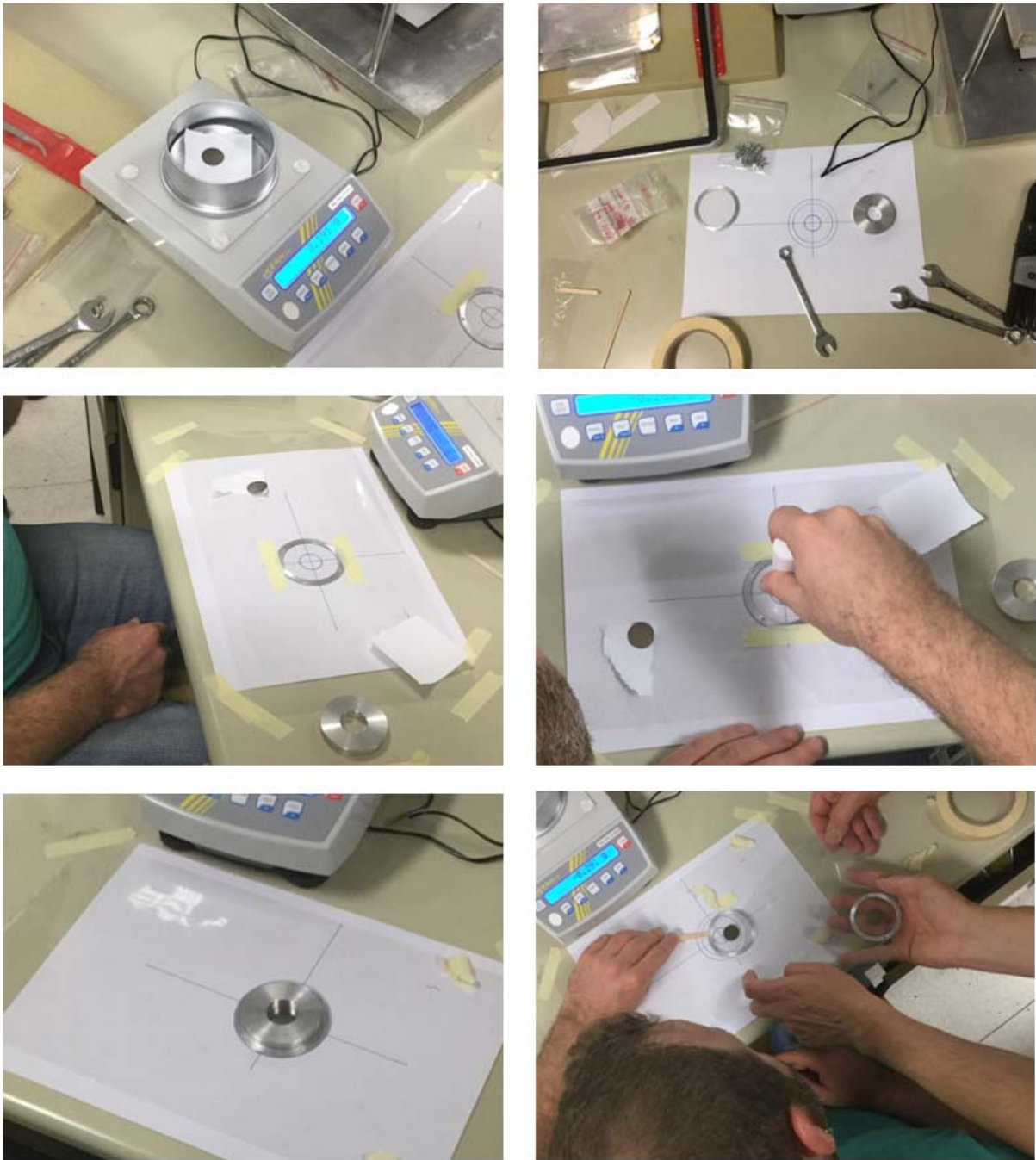


Fig. 5: Preparation of the samples for their arrangement in annular frames to get the correct positioning on the flight path during the experimental campaign.

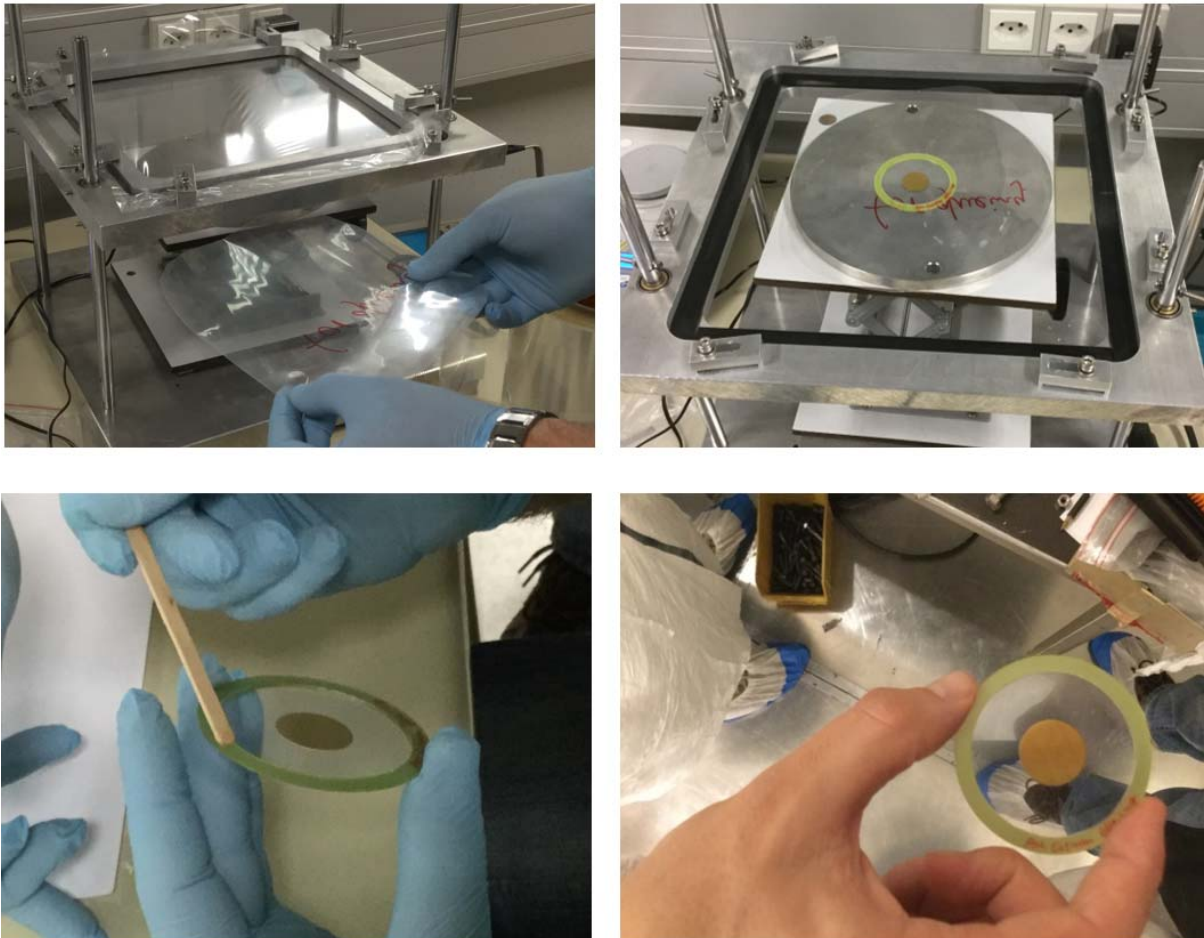


Fig. 6: Clockwise, from the upper picture on the left: operations for glueing the samples between two Mylar foils, in the annular frame to be used for the correct positioning along the beam line. As shown, foils are stretched with a specific tool when they are glued to the frames.

After the completion of the experimental campaign and as soon as possible, a sample isotopic analysis will be performed in order to have additional guarantee that samples features actually comply with the claimed ones.

## 12. The experimental campaign

The experimental campaign for the measurements of the capture cross section of  $^{155}\text{Gd}$  and  $^{157}\text{Gd}$ , by using C6D6 detectors took place between June and July 2016 and lasted for 21 days. During this period, the proton current was monitored with a “pick up” detector operated by the CERN PS team, while neutron beam was monitored in the Experimental Area 1 (EAR1) by means of Silicon Detectors (SiMON) looking at a  $^6\text{LiF}$  foil placed in the beam.

As a rule of thumb, measurements of the neutron capture cross section of a given isotope implies not only the measurement of the sample of interest (Gd odd isotopes in our case), but also a set of auxiliary measurements that are needed for different

purposes: energy calibration, evaluation of different backgrounds, validation of data analysis procedure.

So, in addition to the runs with Gd samples positioned along the beam line and in a plane perpendicular to the beam flight path, further measurements were performed:

- in order to correct for time-dependent background, induced by the neutron beam, measurements were carried out without any sample in the beam. In this case the beam was always on, and the counts are not related to the samples but to the different sources present in the experimental area (Sample out background);
- beam-off, with the aim of characterizing the room background in order to estimate the natural activity and sample activation. This background is sensitive to the sample position, so it is important during the experimental campaign to guarantee for correct positioning and alignment of the samples along the beam line (Beam-off background);
- <sup>197</sup>Au sample, to check with standard cross sections the data analysis procedure;
- carbon sample, because C is an almost pure neutron scatterer, it can be used to evaluate the background due to neutrons scattered from the Gadolinium samples. This type of background can be further investigated through Monte Carlo simulations;
- samples having black resonances at fixed energies (Ag, Co, W, Cd) positioned on the beam line to determine the energy dependence of the background;
- Pb sample, which has a similar atomic number to gadolinium and negligible capture cross section. It was used to estimate the Compton scattering of in-beam gamma rays by the sample.

Details of time allocation for the experiment are summarized in the table below:

<b>Sample</b>	<b>No Filters</b>		<b>With Filters</b>	
	<b>#Protons</b>	<b>Running Time [days]</b>	<b>#Protons</b>	<b>Running Time [days]</b>
<b><sup>155</sup>Gd thin</b>	<b>3.54E+17</b>	<b>4.00</b>	-	-
<b><sup>155</sup>Gd thick</b>	<b>3.29E+17</b>	<b>4.00</b>	<b>4.17E+16</b>	<b>0.50</b>
<b><sup>157</sup>Gd thin</b>	<b>3.96E+17</b>	<b>5.00</b>	-	-
<b><sup>157</sup>Gd thick</b>	<b>4.12E+17</b>	<b>5.00</b>	-	-
<b>Sample Out</b>	<b>1.71E+17</b>	<b>1.00</b>	-	-
<b>Beam Off</b>	-	<b>0.25</b>	-	-

<b>Calibrations</b>	-	<b>0.87</b>	-	-
<sup>nat</sup> Pb	5.75E+16	<b>0.50</b>	-	-
<sup>197</sup> Au	3.63E+16	<b>0.25</b>	-	-
<sup>nat</sup> C	4.71E+16	<b>0.20</b>	-	-
<b>Total</b>	<b>1.81E+18</b>	<b>21.07</b>	<b>4.17E+16</b>	<b>0.50</b>

Table 2. Details of time allocation for the experiment showing the measurements with odd Gd samples and the ancillary ones.

In the table below the main features of the Gd samples are listed.

<b>Isotope</b>	<b>Form</b>	<b>Geometry</b>	<b>Radius [cm]</b>	<b>Isotopic Purity [%]</b>	<b>Weight [mg]</b>	<b>Areal Density [mg/cm<sup>2</sup>]</b>
<sup>157</sup> Gd	metallic	disc	1	88.32	191.6 ±0.1	61.0
<sup>157</sup> Gd	metallic	disc	1	88.32	4.7 ±0.1	1.5
<sup>155</sup> Gd	metallic	disc	1	91.74	100.6 ±0.1	32
<sup>155</sup> Gd	metallic	disc	1	91.74	10.0 ±0.1	3.2

Table 3. Main features of the Gd samples used during the experimental campaign.

The total proton delivery upon the spallation target amounts to 1.81E18 protons. The isotopic enrichment of <sup>155</sup>Gd samples amounts to 91.74%, while that of <sup>157</sup>Gd to 88.32%.

Tables 4 and 5 show the elemental and isotopical compositions of the <sup>157</sup>Gd and <sup>155</sup>Gd samples, respectively, as given by ORNL.



## ASSAY

Element: Gadolinium			SPECTROGRAPHIC ANALYSIS (SSMS)			
Symbol: Gd			Element: ppm	Element: ppm	Element: ppm	Element: ppm
Isotope: 157			Ag: <0.95	t: <0.13	Rh: <0.34	Tc:
Series:			Al: 3.99	In: <0.39	Ru: <0.32	Te: <0.37
Batch: TRAC-2-10			As: <0.22	Ir: <0.9	S: <1	Th: <0.69
ISOTOPIC ANALYSIS			Au: <0.58	K: 6.35	Sb: <0.21	Ti: 2.58
Isotope	Atomic Percent	Precision plus/minus	B: 20.51	Li:	Sc: <1.33	Tl: <0.86
152	<0.01	0.00000	Ba: <6.28	Mg: 7.19	Se: <0.83	U: <0.71
154	0.04	0.01000	Be: <2.96	Mn: 0.81	Si: 17.74	V: <0.15
155	0.29	0.01000	Bi: <0.32	Mo: <0.39	Sn: <0.48	W: 3.8
156	1.68	0.01000	Br: <0.53	N:	Sr: <1.05	Zn: 2.31
157	88.32	0.01000	C: Support	Na: 1.13	Ta: <5.35	Zr: <0.22
158	9.10	0.01000	Ca: 450.53	Nb: <0.09	LANTHANIDES and ACTINIDES	
160	0.57	0.01000	Cb: <0.09	Ni: 7.96		
The limits quoted above are an expression of the precision of this measurement only. The error is estimated at less than 1% from known sources of systematic errors.			Cd: <0.39	O:	Am:	La: 1.37
			Cl: 46.16	Os: <3.48	Bk:	Lu: 2.67
			Co: <0.09	P: 0.15	Ce: 2.4	Md:
			Cr: 1.62	Pu:	Cf:	Nd: <3.5
			Cs: <0.13	Pb: 14.64	Cm:	Np:
			Cu: 9.09	Pd: <0.38	Dy: <1.93	Pr: <0.42
			F: <0.19	Pm:	Er: <2.16	Pu:
			Fe: 11.56	Po:	Es:	Sm: <2.97
			Ga: <0.34	Pt: <17.09	Eu: 02.00	Tb: <0.47
			Ge: <0.3	Ra:	Fm:	Y: 0.26
Internal Use Only Date Entered: 12/30/2009 Last Change: 09/10/2012 09:24:46 AM By: EIH			Hf: <2.84	Rb: <0.39	Gd: Matrix	Yb: 5.36
			Hg: <1.99	Re: <0.88	Ho: <0.49	Tm: 12.5

\* Request No. IDG-0018; Requisition No. 4961; Notebook No. 8003224, pgs 71-73.

\* This analysis reflects enrichment and impurity levels prior to conversion/fabrication.

\* <- No spectrum line visible. Probably absent, definitely less than value given.

\* <T- Present but: less than value given.

\* The spectrographic results reported herein are semi-quantitative estimates and should not be interpreted or construed to be precise quantitative determinations.

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lassay1

Table 4. Chemical and isotopical composition of 157Gd samples.



## ASSAY

Element: Gadolinium	SPECTROGRAPHIC ANALYSIS (SSMS)			
Symbol: Gd	Element: ppm	Element: ppm	Element: ppm	Element: ppm
Isotope: 155	Ag: <50	t	Rh:	Tc:
Series:	Al: <100	In: <500	Ru:	Te:
Batch: 160842	As:	Ir:	S:	Th:
<b>ISOTOPIC ANALYSIS</b>	Au: <500	K: <100	Sb: <500	Ti: <100
Atomic Precision Isotope Percent plus/minus	B: <100	Li: <50	Sc:	Tl:
152 0.03 0.00000	Ba: <100	Mg: <100	Se:	U:
154 0.63 0.02000	Be: <10	Mn: <100	Si: 200	V: <200
155 91.74 0.18000	Bi: <200	Mo: <100	Sn: <200	W: <500
156 5.12 0.18000	Br:	N:	Sr: <100	Zn: <2000
157 1.14 0.00000	C: <100T	Na: <100	Ta: <500	Zr: <200
158 0.94 0.09000	Ca: <500	Nb: <500	<b>LANTHANIDES and ACTINIDES</b>	
160 0.40 0.07000	Cb: <500	Ni: <200	Am: <200	La: <200
The limits quoted above are an expression of the precision of this measurement only. The error is estimated at less than 1% from known sources of systematic errors.	Cd: <500	O:	Bk: <1000	Lu: <50
	Cl:	Os:	Ce: <1000	Md:
	Co: <200	P:	Cf: <500	Nd: <500
	Cr: <100	Pu:	Cm: <500	Np: <500
	Cs: <500	Pb: <200	Dy: <1000	Pr: <500
	Cu: <100	Pd:	Er: <50	Pu: <500
	F:	Pm:	Es: <50	Tb: <500
	Fe: <100	Po: <500	Eu: <50	Y: <50
	Ga: <200	Ra:	Fm: <50	Yb: <20
	Ge: <200	Rb: <200	Gd: M	Tm: <500
Hf: <500	Re:	Ho: <500		
Hg: <500				

\* Symbols: M - major; T - trace; I - interference; < - less than; <= less than/equal to; ~ approximately; nd - not detected.

\* Request Nos. 32582, 34197; Requisition Nos. 12674, 9416; Notebook Nos. A9805, pg 55, A101957, pg 149.

\* This analysis reflects enrichment and impurity levels prior to conversion/fabrication.

\* <- No spectrum line visible. Probably absent, definitely less than value given.

\* <T- Present but less than value given.


\* The spectrographic results reported herein are semi-quantitative estimates and should not be interpreted or construed to be precise quantitative determinations.

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lassay1

Table 5. Chemical and isotopical composition of 155Gd samples.

 <b>Ricerca Sistema Elettrico</b>	<b>Sigla di identificazione</b>	<b>Rev.</b>	<b>Distrib.</b>	<b>Pag.</b>	<b>di</b>
	ADPFISS – LP1 – 100	0	L	23	26

The distribution of protons among all samples irradiated during the experimental campaign is presented in Tab. 3. It is reconstructed from the data files and discarding any corrupted or errata files due to system failures during the data acquisition.

For the capture measurements at n\_TOF, 4 C6D6 scintillation detectors are used, in order to reveal signals produced by the prompt gamma rays following each capture event in the sample positioned along the beam line.

C6D6 detectors were specially optimized in order to exhibit as low neutron sensitivity as possible, which is a specific requirement to the low background neutron capture measurements. The detectors used during the experimental campaign, are positioned in a plane perpendicular to the beam line, few centimeters downstream of the sample, thus reducing the effect of scattered in-beam gamma rays, while providing the reasonable geometric efficiency for detecting the gamma-rays due to the deexcitation of the compound nucleus.

The experimental setup for the measurements consisted of an array of four C6D6 scintillators opposite each other at 45 degree with respect to the beam. In fig. 7 pictures of the experimental setup during the measurement campaign are shown. In the picture at the upper left hand corner it is possible to see the BaF<sub>2</sub> crystals which the Total Absorption Calorimeter consists of.

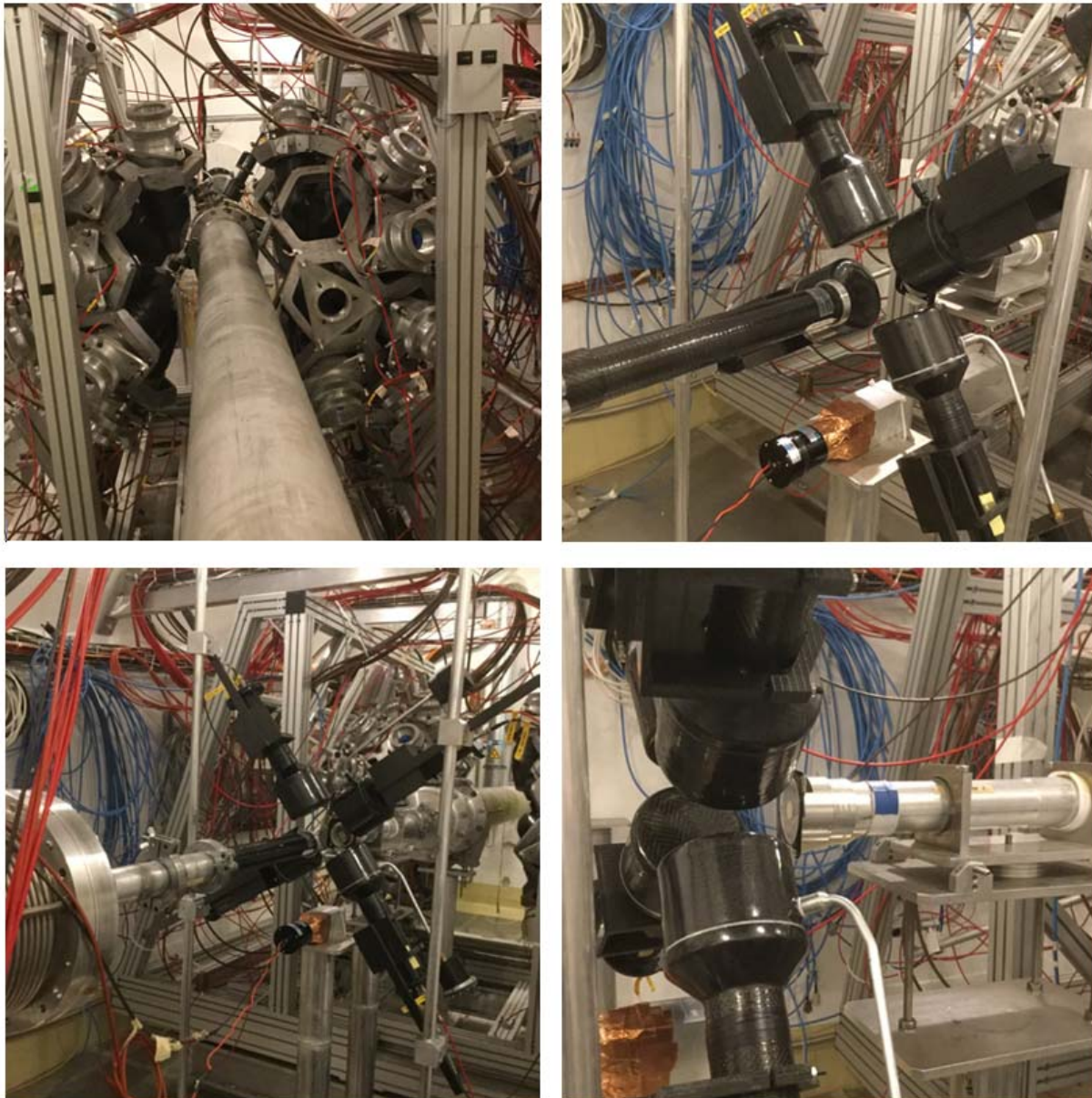



Fig. 7. Experimental setup in EAR1 during the Gd campaign.

The aim of the data analysis is to calculate the energy-dependent capture yield of  $^{155}\text{Gd}(n, \gamma)$  and  $^{157}\text{Gd}(n, \gamma)$  reactions.

From the capture yield the pointwise capture cross section and the resonance parameters can then be extracted.

 <b>Ricerca Sistema Elettrico</b>	<b>Sigla di identificazione</b>	<b>Rev.</b>	<b>Distrib.</b>	<b>Pag.</b>	<b>di</b>
	ADPFISS – LP1 – 100	0	L	25	26

### 13. Conclusions

Based on the outcomes of previous works [1], [2] on the justification for new evaluations of the neutron capture cross sections of Gadolinium odd isotopes, new experimental measurements were carried out at the European Organization for Nuclear Research (CERN) in the framework of the n\_TOF Collaboration.

The experimental campaign was performed in the Experimental Area 1 (EAR1) of the n\_TOF facility at CERN by means of H-free organic scintillators (C6D6). Measurements, performed with the attendance also of the ENEA researchers, took place between June and July 2016 and lasted for 21 days using Gd samples acquired from ORNL.

Data analysis of the  $^{157}\text{Gd}$  and  $^{155}\text{Gd}$  neutron capture cross section measurements is currently underway with the ENEA contribution and is scheduled for completion before the end of 2017.

In order to get a reference value in the thermal region, also thermal cross section measurements at the Budapest Research Reactor was performed in early October.

Moreover, a request for beamtime at IRMM (Gelina) facility was submitted and accepted with the endorsement from JRC-Geel to compare new cross sections with transmission measurements on the same samples.

At the end of the analysis it will be possible to get new values for the Gd odd isotopes capture cross sections which could lead to new assessments about the impact on the neutron safety for the II and III generations of fission reactors.

After the nTOF data analysis is completed, new evaluations for the cross sections of interest will be done, and the validation process will be performed making recourse to the relevant ICSBEP benchmarks.


The related work will be fully detailed in later dedicated Reports.

### 14. Acknowledgments

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We also acknowledge the contribution of the other members of the ENEA team in n\_TOF - Alberto Mengoni, Sergio Lo Meo (spokesperson of the experiment for ENEA), Giulia Clai and Antonio Guglielmelli.



 <b>Ricerca Sistema Elettrico</b>	<b>Sigla di identificazione</b>	<b>Rev.</b>	<b>Distrib.</b>	<b>Pag.</b>	<b>di</b>
	ADPFISS – LP1 – 100	0	L	26	26

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