Towards the next generation of detectors for *n*-γ capture reactions at n_TOF (CERN)

Agatino Musumarra [1,2*], *Nikolas* Patronis [3], *Cristian* Massimi[4,5], *Maria Grazia* Pellegriti [2], and *Dimitrios* Papanikolaou[3] for the n_TOF collaboration

[1]Department of Physics and Astronomy, University of Catania, Italy
[2]INFN Sezione di Catania, Istituto Nazionale di Fisica Nucleare, Catania, Italy
[3]Department of Physics, University of Ioannina, Greece
[4]Department of Physics and Astronomy, University of Bologna, Italy
[5]INFN Sezione di Bologna, Istituto Nazionale di Fisica Nucleare, Bologna, Italy

Abstract. Benzene-d6 has been the scintillation material of choice for performing most of the neutron-capture cross-section measurements at CERN n_TOF for more than 20 years. Recently a revamping of solid-state Stilbene scintillators has been justified by their good timing performances, lack of toxicity and excellent neutron-gamma discrimination by Pulse-Shape Analysis. Consequently, several measurements were recently performed at the n_TOF facility at CERN and INFN-Catania to characterize their performance. We benchmarked both regular and deuterated stilbene scintillation crystals in EAR2@CERN. Based on these results, we are developing and designing a new generation of detectors with enhanced detection sensitivity and improved safety and maintenance conditions.

1 The n_TOF facility at CERN

The neutron spallation facility n_TOF was installed at CERN in the early 2000s, following an original idea by Carlo Rubbia [1]. The neutron beam is produced by the 20 GeV/c proton beam accelerated by the CERN PS impinging on a lead target. The neutron energy at the sample location is precisely determined by using Time of Flight (TOF) over a broad energy range, from meV to GeV (more than 10 decades). The facility develops around three complementary experimental areas: the first one has been installed at the end of a horizontal flight path 185 m long (EAR1) [2] for optimal *n*-energy resolution, while the second beam line, with a vertical flight path of 20 m (EAR2) [3], was conceived to get a decisive boost in neutron flux. Finally, a third experimental area (NEAR) was recently installed, close to the spallation target, for performing cross-section measurements via the activation technique and to study the response of materials and electronics to the intense neutron-gamma field [4]. The drawing of EAR1 and EAR2, with respect to the spallation target, is depicted in Fig. 1.

Since its inception, the facility has been devoted to studying neutron-induced reactions involved in a wide variety of research fields.

Fundamental Nuclear Physics and Astrophysics, ranging from primordial nucleosynthesis, stellar and explosive nucleosynthesis to nuclear structure and fission dynamics.

Applications and nuclear power plant technology, including nuclear waste transmutation, accelerator driven systems and nuclear fuel cycle investigations.

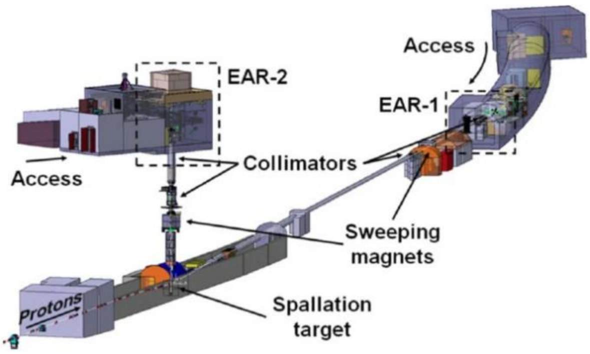

Fig.1. Schematic representation of the n_TOF facility at CERN.

2 Tools for neutron capture reactions

2.1 Evolution of the C_6D_6 detection systems

Among the physics cases supported by the facility, neutron capture reactions (*n,*γ), (*n,cp*) and (*n,fission*) play an important role, since they are involved both in Nuclear Physics and Astrophysics (*e.g.* s-process nucleosynthesis and nuclear spectroscopy) and nuclear technology (*e.g.* actinides transmutation), so that in many cases the experiment can provide useful data both for fundamental physics and applications [5,16]. Since

* Corresponding author: musumarra@lns.infn.it

© The Authors, published by EDP Sciences. This is an open access article distributed under the terms of the Creative Commons Attribution License 4.0 (https://creativecommons.org/licenses/by/4.0/).

the beginning of the experimental study of (*n,γ*) capture reactions, the chosen method for detecting secondary γ-rays was deuterated benzene (C_6D_6). For more than 20 years this hazardous liquid scintillator has been arranged in several geometrical configurations around the target sample with the main task of combining a large solid angle and intrinsic efficiency with a low background and neutron sensitivity [6]. A typical arrangement of a C_6D_6 detector simply consists of a cylindrical tank of benzene scintillator coupled to a photomultiplier and a voltage divider. At n_TOF the tank and the detector case have been assembled by using Carbon fibre (INFN-Legnaro) [7] or Aluminium (Bicron), as shown in Fig. 2. In this setup the need for reducing and optimizing the total mass surrounding the scintillator (thus minimizing background) clashes with the mandatory safety requirements for containing and sealing the benzene scintillator. Recently, a crucial development occurred when it was demonstrated that a closed geometry of small C_6D_6 (referred to as sTED) modules, placed few centimetres far from the target, could provide a good detection efficiency with a significant improvement in signal to noise (S/N) ratio in EAR2 [8]. The sTED modules are now routinely used in EAR2 for measuring (*n,γ*) capture reactions, sometimes in combination with the reference, the standard C_6D_6 detectors.

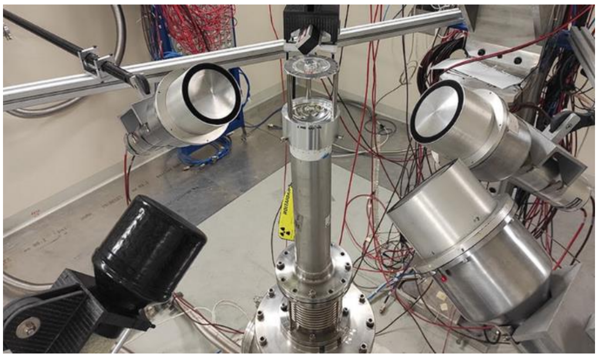

Fig. 2. One Legnaro (on the left) and three Bicron C_6D_6 detectors surrounding the target in EAR2.

2.2 A solid-state option for the next generation *n*-capture setup at n_TOF: trans-Stilbene

The idea of using solid organic scintillators as a suitable alternative to liquid materials is not new. In fact, the scintillation properties of trans-Stilbene have been extensively investigated since the 60s [9]. The limitations arise mainly from the technology for growing high purity crystals, which often hinder to reach large detection volumes, and the anisotropy response to impinging radiation. Recently, thanks to the efforts of the Lawrence Livermore National Labs (LLNL) [10], large, rapidly grown, standard and deuterated Stilbene crystals have been produced. At the same time, they have been made available to the market by Inrad Optics [11] and Proteus [12]. On the other hand, it has been verified that the crystal anisotropy concerns only neutrons and heavy-ions and does not affect γ-ray response [13].

Implementing detectors using trans-Stilbene organic scintillator provides several advantages (see Table 1): it is a solid and non-toxic material, has a higher density (15% more than benzene), and it offers excellent γ/n discrimination capability via Pulse Shape Analysis (PSA). Consequently, the scintillator housing mass can be minimized. This reduction in the total surrounding mass, along with the γ/n discrimination abilities, allows for minimal neutron sensitivity, when detecting neutron-capture emitted gamma rays. Simultaneously, the same detector can be used for neutron elastic and neutron inelastic scattering studies at neutron energies ($E_n > 100$ *keV*).

According to the scintillation spectral bandwidth, Stilbene can be coupled both with Photomultipliers Tubes (PMT) and Silicon-Photomultipliers (SiPM). The large scintillation light-yield (50-60 % with respect to anthracene [13]) is a key parameter to optimize a smart combination of PMT and active voltage divider, as described in the following section.

Table 1. Trans-Stilbene properties from [11].

Density	1.15 g/cm^3
Refractive Indices (*at 589 nm*)	1.703, 1.724, 1.844
Maximum Emission Wavelength	380 nm
Decay Time (*Prompt*)	4.3 ns

2.3 The PSTIL design at n_TOF, and its benchmark.

The first prototype, hereafter referred to as PSTIL, for in-beam testing at n_TOF was a simple, and very performing assembly. This was the first protonated Stilbene detector ever used at n_TOF (June-September 2022). The detector readout components are shown in Fig. 3. The PMT was coupled to a cylindrical trans-Stilbene crystal 3.2 cm in diameter by 1 cm length provided by Inrad Optics.

Fig. 3. The PSTIL detector readout components: HAMAMATSU PMT model R1924A (borosilicate window) and the Sens-Tech PS1807 DC-DC converter (with active voltage divider).

The choice of the HAMAMATSU R1924A PMT (Bialkali photocathode and borosilicate window) in combination with the PS1807 active voltage divider allowed us for maintaining a low PMT gain and high-rate capability. In fact, the HV voltage applied was 600 V, close to the minimum working voltage of 500 V. The optimized PMT gain, and the small crystal size were the key factors to reduce the γ-flash component during the test and sustain the high-rate induced by the high neutron flux in EAR2. Thanks to this setup we were able to exploit all the excellent scintillator properties: fast timing (fall-time 3 ns), high rate (pulse-width 20 ns) and the PSD capability, preserving the pulse tail (130 ns slow component).

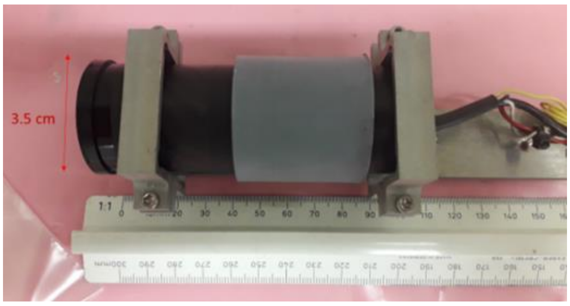

Fig. 4. The first n_TOF Stilbene detector including, in a compact design, a 3.2 cm diameter by 1 cm length cylindrical standard Stilbene crystal by Inrad Optics. Carbon fibre was used for the case.

2.4 First experimental results

The PSTIL prototype was tested at the n_TOF EAR2 in combination with the sTED modules and the bulky C_6D_6 reference detectors for comparison. The detector was placed at 90° degrees with respect to the beam direction, 6 cm from the target sample as shown in Fig. 5.

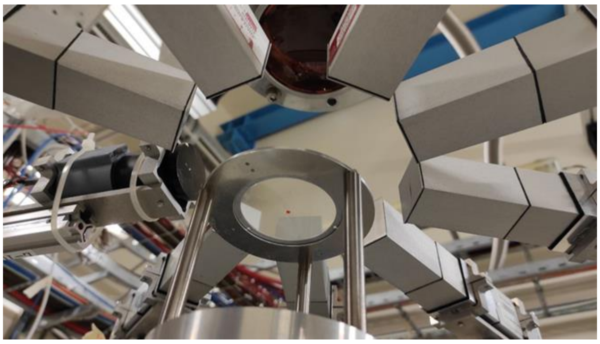

Fig. 5. The PSTIL prototype installed in EAR2, placed at the same geometrical position with respect the eight sTED modules during the Mo*(n,γ)* measurement campaign performed in 2022 [5].

All the detection modules were connected to the n_TOF DAQ digitizers using the same cable routing and patch panel. The system was calibrated by using standard 137-Cs, 207-Bi, 88-Y and Am-Be *γ* and *γ-n* sources. There was no need to optimize grounding, as baseline noise was very low (1 mV RMS), the PMT polarization voltage was the nominal one.
The performance was outstanding, as confirmed by the time-of-flight (TOF) spectra for the Au*(n,γ)* benchmark *n*-capture reaction. As shown in Fig. 6 the overall detection performances, specifically the signal to noise ratio, is on a par with the sTED detectors.
The experimental evidence, once again, confirms that a compact and segmented array of small detection modules can outperform larger and heavier scintillation systems, when placed in a proper geometrical arrangement around the target. A tenfold improvement in the *S/N* ratio has been obtained by PSTIL and sTED with respect to the classical C_6D_6 setup.

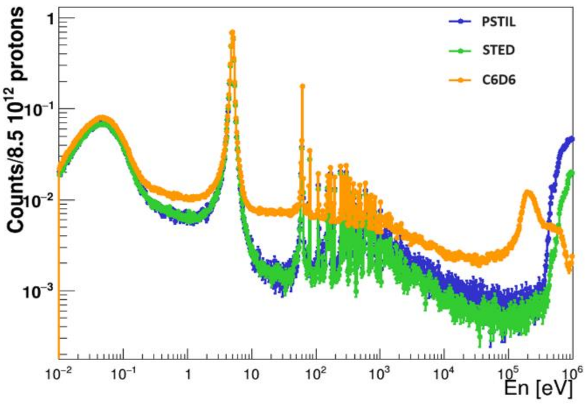

Fig. 6. Normalized E_n-TOF spectra obtained for the Au*(n,γ)* capture reaction in EAR2 (courtesy of J. Balibrea). The comparison shows similar performances obtained by PSTIL with respect to the sTED modules.

The stability of the PSTIL performance was verified in a second run performed in EAR2, as reported in Fig.7, which focuses on the energy range of the Au saturated resonance (E_n =5 eV).

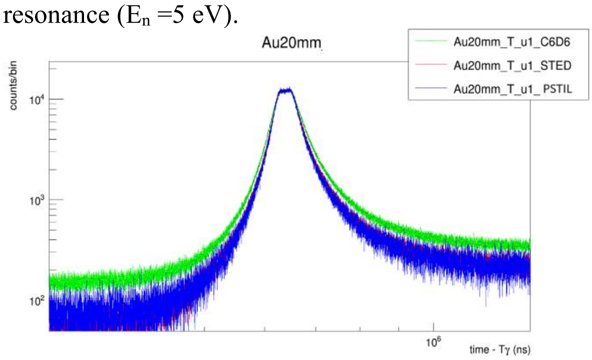

Fig. 7. TOF spectra obtained for the Au*(n,γ)* capture reaction saturated resonance in EAR2 (courtesy of R. Mucciola).

Finally, it is worthwhile to stress that these results were obtained using a lightweight and low-cost setup where a state-of-the-art DC-DC converter avoids any HV power supply and cabling. In addition, the low power requirement (5 V, 50 mA) allows for peculiar setup flexibility, opening the possibility of using the detector in harsh environments (including neutron detection in space).

3 The first trans-Stilbene operational detectors array at n_TOF

The result obtained in 2022 by this detector design served as the basis for building a Stilbene array consisting of eight detectors. Some optimizations were implemented on the single module, maintaining the same concept and components. In the final configuration a full carbon fibre case was used. Additionally, a 1-inch by 1-inch cylindrical Stilbene crystal was coupled to the HAMAMATSU PM model R7378A (synthetic silica version of the R1924A). A 50-micron Al window was placed in front of the scintillator. The final detection module is shown in Fig. 8. The assessment of the performances of the first batch of four detectors was performed in EAR2. More specifically, a first Letter of Intent (LoI) was presented and accepted by the INTC scientific committee of CERN (INTC-2023-034; INTC-I-254) [14].

Fig. 8. The final PSTIL module with the new carbon fibre case. The HAMAMATSU PMT R7378A was coupled to the 1' x 1' cylindrical Stilbene crystal. Four trans-Stilbene crystals were provided by Inrad Optics [11] and further four crystals by Proteus [12].

The experiment was performed in May 2023. During the run, we had the opportunity to validate the detector performance in terms of neutron detection by using a carbon target and arranging the four modules at 90° and 135° with respect to the beam direction. The EAR2 setup is shown in Fig.9. Detection redundancy at the same scattering angles was crucial in order to cross-check the angular distributions for $^{12}C(n,n)$ and $^{12}C(n,n')$ elastic and inelastic scattering channels in the neutron energy range 1 MeV$<E_n<$10 MeV.

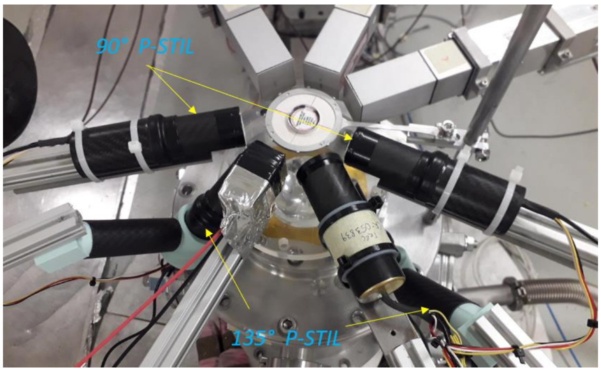

Fig. 9. EAR 2 setup, four PSTIL modules were used, placed at 90° and 135°.

The detectors were able to sustain the high instantaneous *n*-flux in EAR2 (despite a large pile-up component being present). Consequently, the E_n-Tof spectra were clearly modulated by the angular distribution according to the different elastic and inelastic reaction channels. The analysis is in progress, and based on the preliminary results a further run was proposed to investigate the response of PSTIL to (n,n) and (n,n') reaction channels: Letter of Intent CERN-INTC-2024-028;INTC-I-274 [15]. The run is planned in EAR1 in October 2024 with the same carbon target, in order to improve the energy resolution and decrease the pile-up probability while investigating the 1-20 MeV energy range.

4 Perspectives

Based on the experimental characterization we have confirmed that trans-Stilbene is a valuable alternative to deuterated benzene (C_6D_6) for designing a new detector array suitable for (n,γ) reactions measurements at n_TOF. Moreover, this solid material improves safety conditions, simplifies the setup and increases the intrinsic detection efficiency. The capability for state-of-the-art γ-n discrimination extends the use of non-deuterated stilbene scintillator to (n,n) and (n,n') elastic and inelastic scattering measurements at large neutron energies ($E_n>100$ KeV). An array of eight modules is currently available at n_TOF CERN. This array will be utilized before and after the LS3 period to complement the actual measurements, as also stated in [16] and in combination with other detection systems (*e.g.* $LaBr_3$) for conducting inelastic measurements in an unexplored neutron energy range at n_TOF.

References

1. C. Rubbia at al., A high resolution spallation driven facility at the CERN-PS to measure neutron cross sections in the interval from 1 eV to 250 MeV: a relative performance assessment. Addendum to CERN-LHC-98-002-EET(1998). https://cds.cern.ch/record/363828
2. C. Guerrero et al., Performance of the neutron time-of-flight facility n_TOF at CERN. Eur. Phys. Jour. A **49**, 27 (2013).https://doi.org/10.1140/epja/i2013-13027-6
3. C. Weiß et al., The new vertical neutron beam line at the CERN n_TOF facility design and outlook on the performance. Nucl. Inst. Methods A **799**, 90 (2015).https://doi.org/10.1016/j.nima.2015.07.027
4. M.E. Stamati et al., The n_TOF NEAR Station Commissioning and first physics case. ND2022 Conference, EPJ Web of Conferences **284**, 06009 (2023).https://doi.org/10.1051/epjconf/202328406009
5. C. Massimi, Measurement of 94,95,96-Mo(n,γ) relevant to Astrophysics and Nuclear Technology. Proposal CERN-INTC-2020-052; INTC-P-569 (2020). https://cds.cern.ch/record/2730968
6. R. Plag et al., An optimized C6D6 detector for studies of resonance-dominated (n,γ) cross-sections. Nucl. Inst. Methods A **496**, 425 (2003). https://doi.org/10.1016/S0168-9002(02)01749-7
7. P. Mastinu et al., New C6D6 detectors: reduced neutron sensitivity and improved safety. Internal note n_TOF-PUB-2013-002; CERN-n_TOF-PUB-2013-002. https://cds.cern.ch/record/1558147
8. V. Alcayne, et al., A segmented total energy detector (sTED) for (n,γ) cross section measurements at n_TOF EAR2. ND2022 Conference, EPJ Web of Conferences **284**, 01043 (2023).https://doi.org/10.1051/epjconf/202328401043

9. F. D. Brooks, A scintillation counter with neutron and gamma-ray discriminators. Nucl. Instr. Meth. **4** (1959) 151.https://doi.org/10.1016/0029-554X(59)90067-9
10. N. Zaitseva et al., Scintillation properties of solution-grown trans-stilbene single crystals. Nucl. Inst. Methods A **789** (2015) 8. https://doi.org/10.1016/j.nima.2015.03.090
11. Inrad Optics, 181 Legrand Avenue, Northvale, NJ 07647.https://www.inradoptics.com/
12. Proteus, 120 Senlac Hills Dr, Chagrin Falls, OH 44022.https://proteus-pp.com/
13. A. Di Chicco et al., Gamma-response characterization of a solution-grown stilbene based detector assembly in the 59 keV–4.44 MeV energy range; an alternative low-resolution gamma spectrometer. Nucl. Inst. Methods A **1034** (2022) 166740.https://doi.org/10.1016/j.nima.2022.166740
14. J. Balibrea and A. Musumarra, Development of new solid-state total-energy detectors for neutron-capture measurements at CERN n_TOF. Proposal CERN-INTC-2023-034 ; INTC-I-254.http://cds.cern.ch/record/2856350/files/INTC-I-254.pdf
15. M.G. Pellegriti and N. Sahoo, Response of stilbene detectors to (n,n) and (n,n') reaction channels in TOF experiments. Letter of Intent CERN-INTC-2024-028 ; INTC-I-274.https://cds.cern.ch/record/2894839/files/INTC-I-274.pdf
16. C. Massimi, Study of n+63,65-Cu reactions and their relevance for nuclear technologies and Astrophysics. Proposal CERN-INTC-2024-006; INTC-P-689.https://cds.cern.ch/record/2886127